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(54) **SUBSTITUTED PYRAZOLE DERIVATIVE AND AGROHORTICULTURAL BACTERICIDE.**

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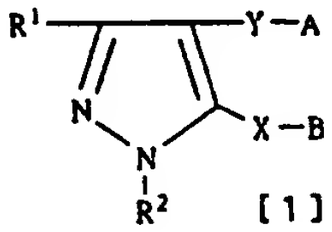
(57) A nov l substituted pyrazole derivative represent d by general formula (1) and an agrohorticultural bactericide containing the sam , wherein R^1 represents hydrogen, halogen, alkyl, alkoxy, alkylthio or haloalkyl; R^2 represents hydrogen, alkyl, haloalkyl, optionally substituted phenylalkyl, $-COR^6$ or $-SO_2R^7$; X represents $-S(O)_{0-2}$, $-NR^3-$, -CO- or $-CR^4R^5-$; Y represents -O- or $-S(O)_{0-2}$; A represents optionally substituted phenyl or heterocyclic group; and B represents optionally substituted pyridyl, diazinyl, 1, 3, 6- or 1, 3, 4- triazinyl or thiazolyl. The above compound is useful as an agrohorticultural bactericide, because it has an excellent agrohorticultural bactericidal action and is free from chemical injury against useful crops.



[1]

TECHNICAL FIELD

The present invention relates to novel pyrazole derivatives and fungicides for agricultural and horticultural use containing the derivative(s) as an active ingredient.

BACKGROUND ART

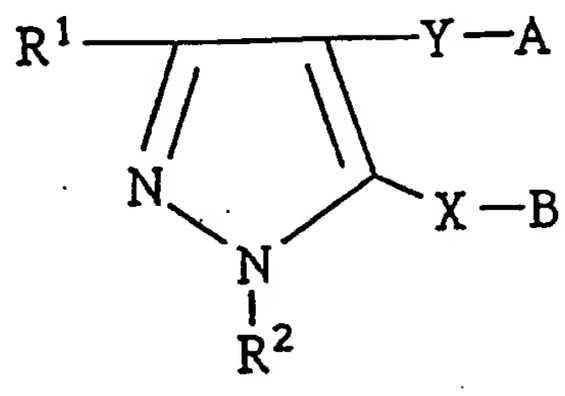
Various fungicides have heretofore been developed, but the potency of them could not be said to be always satisfactory due to appearance of resistant strains and other reasons.

Japanese Patent Application Laid-Open No. 1-125379 mentions that pyrazole derivatives of certain kinds have fungicidal activity.

The compounds as disclosed in the laid-open specification are not still satisfactory with respect to the potency, the residual effect and the phytotoxicity. Therefore, development of fungicides for agricultural and horticultural use, which are further more useful to plant diseases, is desired.

DESCRIPTION OF THE INVENTION

In view of the situation mentioned above, the present inventors repeatedly made various investigations so as to develop compounds having excellent fungicidal activity and, as a result, have found that substituted pyrazole derivatives of the following general formula (I) have excellent fungicidal activity. On the basis of the finding, they have achieved the present invention. Specifically, the present invention relates to substituted pyrazole derivatives of a general formula [1]:



[1]

where R^1 represents a hydrogen atom, a halogen atom, an alkyl group, an alkoxy group, an alkylthio group or a haloalkyl group;

R^2 represents a hydrogen atom, an alkyl group, a haloalkyl group, an unsubstituted or substituted phenylalkyl group, $-COR^6$ or $-SO_2R^7$;

X represents -S-, -SO-, $-SO_2-$, $-N(R^3)-$, -CO- or $-C(R^4)(R^5)-$;

R^3 represents a hydrogen atom, an alkyl group, a haloalkyl group, an alkenyl group, an alkynyl group, an alkoxyalkyl group, a cyanoalkyl group, an alkylcarbonylalkyl group, an alkoxycarbonylalkyl group, a nitroso group, an amino group, an unsubstituted or substituted phenylalkyl group, $-COR^6$ or $-SO_2R^7$;

R^4 and R^5 independently represent a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkenyl group, an alkynyl group or $-OR^8$;

R^8 represents a hydrogen atom, an alkyl group, a haloalkyl group, an alkenyl group, an alkynyl group, an alkoxyalkyl group, a cyanoalkyl group, an alkylcarbonylalkyl group, an alkoxycarbonylalkyl group, an unsubstituted or substituted phenylalkyl group, $-COR^6$ or $-SO_2R^7$;

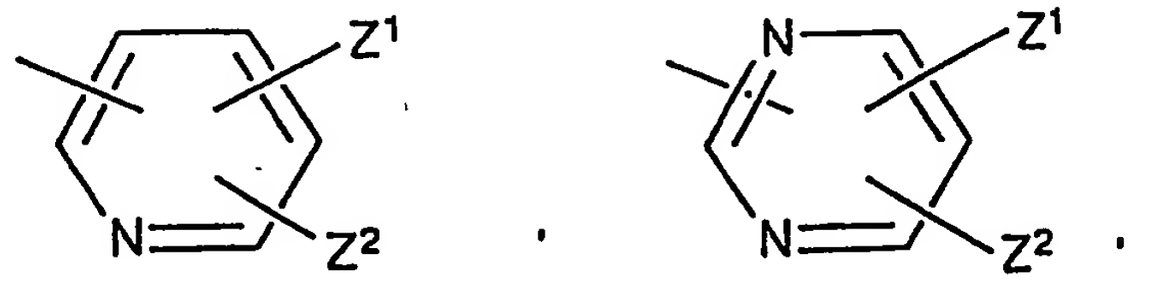
R^6 represents a hydrogen atom, an alkyl group, a haloalkyl group, an unsubstituted or substituted phenyl group, an unsubstituted or substituted phenylalkyl group, an alkoxy group or

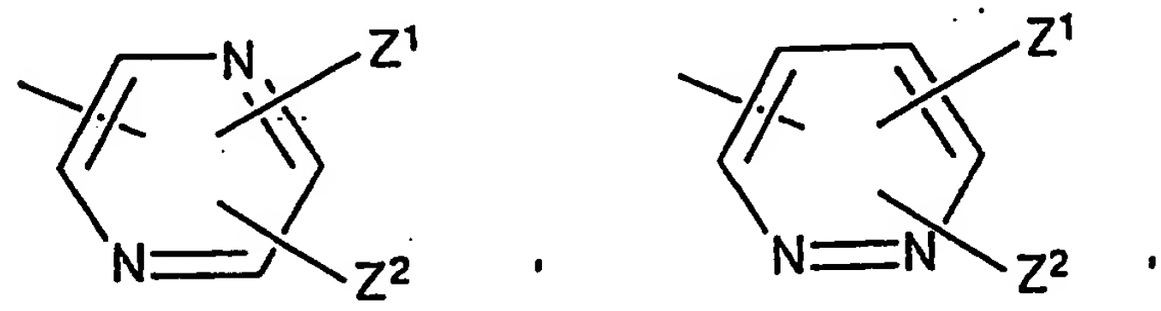
$$-N<\begin{matrix}R^9\\R^{10}\end{matrix}\quad;$$

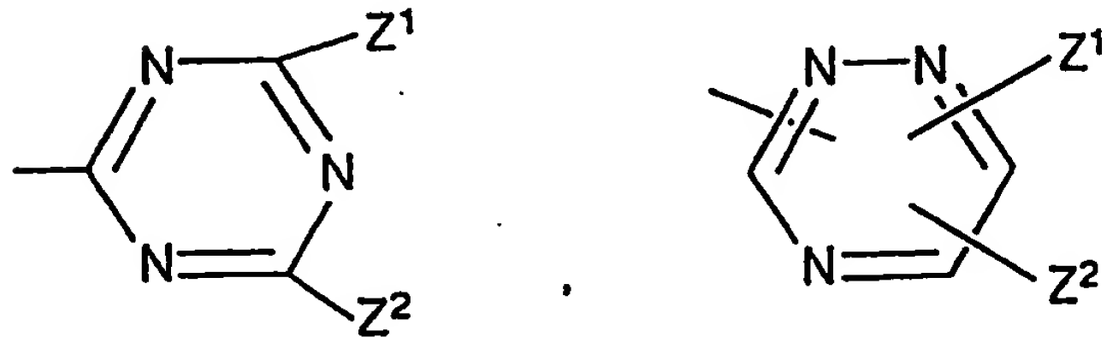
R^7 represents an alkyl group, a haloalkyl group, an unsubstituted or substituted phenyl group or

;

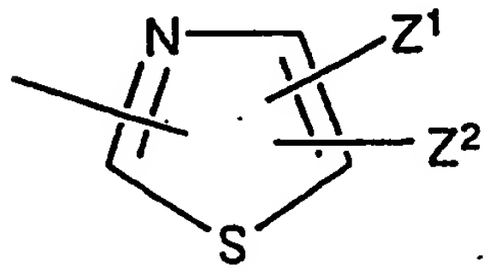
R⁹ and R¹⁰ independently represent a hydrogen atom, an alkyl group or an unsubstituted or substituted phenyl group;
Y represents an oxygen atom, -S-, -SO-, or -SO_2-;
A represents an unsubstituted or substituted phenyl group or an unsubstituted or substituted heterocyclic group;
B represents







or



;

Z^1 and Z^2 independently represent a hydrogen atom, a halogen atom, an alkyl group, an alkoxy group or a haloalkyl group;
and also to fungicides for agricultural and horticultural use containing th derivative(s) as an activ ingredient.

Next, compounds of formula [1] of the present invention are shown in Table 1 and Table 2. However, compounds of the present invention are not restricted to only them.

The compound number is referred to in the following description herein. In the tables, Ph indicates a phenyl group, i indicates iso-, and t indicates tertiary.

Table 1

In compounds of:

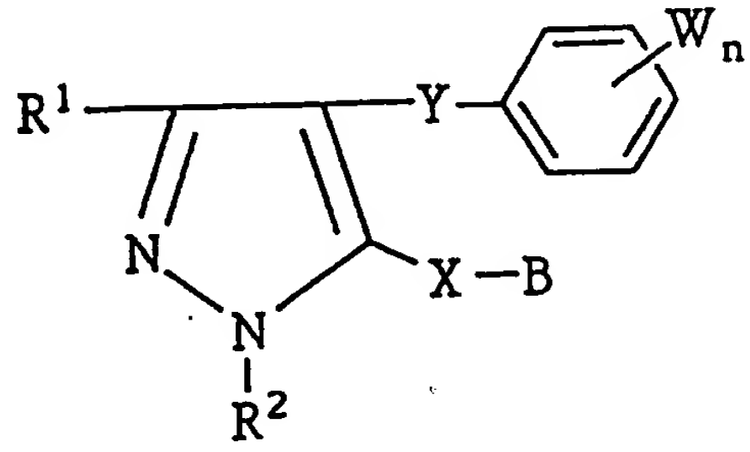


Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
1	H	CH_3	S	S	H	B 1
2	CH_3	CH_3	S	S	H	B 1
3	CF_3	CH_3	S	S	H	B 1
4	H	CH_3	S	S	4-Cl	B 1
5	CH_3	CH_3	S	S	4-Cl	B 1
6	CF_3	CH_3	S	S	4-Cl	B 1
7	CH_3	CH_3	S	S	$4-CH_3$	B 1
8	CF_3	CH_3	S	S	$4-CH_3$	B 1
9	CH_3	H	S	S	4-Cl	B 1
10	CH_3	H	S	S	$4-CH_3$	B 1
11	CH_3	CF_3	S	S	4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
12	CH_3	CF_3	S	S	4-CH_3	B 1
13	CH_3	CH_3	S	S	4-CF_3	B 1
14	CH_3	CH_3	S	S	4-Br	B 1
15	CH_3	CH_3	S	S	4-NO_2	B 1
16	CH_3	CH_3	S	S	4-OCH_3	B 1
17	CH_3	CH_3	S	S	4-C_2H_5	B 1
18	CH_3	CH_3	S	S	2-Cl	B 1
19	CH_3	CH_3	S	S	2-Cl, 4-Cl	B 1
20	CH_3	CH_3	S	S	2-Cl, 4-CH_3	B 1
21	CH_3	CH_3	S	S	4-Cl	B 2
22	CF_3	CH_3	S	S	4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
23	CH_3	CH_3	S	S	4-CH_3	B 2
24	CF_3	CH_3	S	S	4-CH_3	B 2
25	CH_3	CH_3	S	S	2-Cl, 4-Cl	B 2
26	CH_3	CH_3	S	S	2-Cl, 4-CH_3	B 2
27	CH_3	CH_3	S	O	4-Cl	B 2
28	CH_3	CH_3	S	O	4-CH_3	B 2
29	CF_3	CH_3	S	O	4-CH_3	B 2
30	CH_3	CH_3	S	O	3-Cl, 4-CH_3	B 2
31	C_2H_5	CH_3	S	S	H	B 1
32	C_2H_5	CH_3	S	S	4-Cl	B 1

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
33	C_2H_5	CH_3	S	S	4-CH_3	B 1
34	C_2H_5	CH_3	S	S	4-OCH_3	B 1
35	C_2H_5	CH_3	S	S	3-CF_3	B 1
36	C_2H_5	CH_3	S	S	2-Cl, 4-Cl	B 1
37	C_2H_5	CH_3	S	S	3-Cl, 4-Cl	B 1
38	C_2H_5	CH_3	S	S	2-Cl, 4-CH_3	B 1
39	i-C_3H_7	CH_3	S	S	4-Cl	B 1
40	i-C_3H_7	CH_3	S	S	4-CH_3	B 1
41	i-C_3H_7	CH_3	S	S	4-OCH_3	B 1
42	t-C_4H_9	CH_3	S	S	4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
43	t-C_4H_9	CH_3	S	S	4-CH_3	B 1
44	t-C_4H_9	CH_3	S	S	4-OCH_3	B 1
45	CH_3	C_2H_5	S	S	4-Cl	B 1
46	CH_3	C_3H_7	S	S	4-Cl	B 1
47	CH_3	CH_3	NH	S	H	B 1
48	CF_3	CH_3	NH	S	H	B 1
49	CH_3	CH_3	NH	S	4-Cl	B 1
50	CF_3	CH_3	NH	S	4-Cl	B 1
51	H	CH_3	NH	S	4-Cl	B 1
52	CH_3	CH_3	NH	S	4-CH_3	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
53	CF_3	CH_3	NH	S	4-CH_3	B 1
54	H	CH_3	NH	S	4-CH_3	B 1
55	CH_3	CH_3	NH	S	4-F	B 1
56	CH_3	CH_3	NH	S	4-Br	B 1
57	CH_3	CH_3	NH	S	4-I	B 1
58	CH_3	CH_3	NH	S	2-F	B 1
59	CH_3	CH_3	NH	S	2-Cl	B 1
60	CH_3	CH_3	NH	S	2-Br	B 1
61	CH_3	CH_3	NH	S	2-I	B 1
62	CH_3	CH_3	NH	S	3-F	B 1

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
63	CH_3	CH_3	NH	S	3-Cl	B 1
64	CH_3	CH_3	NH	S	3-Br	B 1
65	CH_3	CH_3	NH	S	3-I	B 1
66	CH_3	CH_3	NH	S	2-OCH_3	B 1
67	CH_3	CH_3	NH	S	3-OCH_3	B 1
68	CH_3	CH_3	NH	S	4-OCH_3	B 1
69	CH_3	CH_3	NH	S	2-CF_3	B 1
70	CH_3	CH_3	NH	S	3-CF_3	B 1
71	CH_3	CH_3	NH	S	4-CF_3	B 1
72	CH_3	CH_3	NH	S	4-C_2H_5	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
73	CH_3	CH_3	NH	S	4-C_3H_7	B 1
74	CH_3	CH_3	NH	S	4-C_4H_9	B 1
75	CH_3	CH_3	NH	S	4-i-C_3H_7	B 1
76	CH_3	CH_3	NH	S	4-t-C_4H_9	B 1
77	CH_3	CH_3	NH	S	2-OCF_3	B 1
78	CH_3	CH_3	NH	S	3-OCF_3	B 1
79	CH_3	CH_3	NH	S	4-OCF_3	B 1
80	CH_3	CH_3	NH	S	3-NO_2	B 1
81	CH_3	CH_3	NH	S	4-NO_2	B 1
82	CH_3	CH_3	NH	S	3-NH_2	B 1

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
83	CH_3	CH_3	NH	S	4-NH_2	B 1
84	CH_3	CH_3	NH	S	4-$NHCOCH_3$	B 1
85	CH_3	CH_3	NH	S	4-$NHSO_2CH_3$	B 1
86	CH_3	CH_3	NH	S	4-$NHCOCF_3$	B 1
87	CH_3	CH_3	NH	S	4-$NHSO_2CF_3$	B 1
88	CH_3	CH_3	NH	S	4-Ph	B 1
89	CH_3	CH_3	NH	S	4-OPh	B 1
90	CH_3	CH_3	NH	S	4-OCF_2CF_2H	B 1
91	CH_3	CH_3	NH	S	4-$COCH_3$	B 1
92	CH_3	CH_3	NH	S	4-NHCOPh	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
93	CH_3	CH_3	NH	S	4-$NHCOOCH_3$	B 1
94	CH_3	CH_3	NH	S	4-$NHCON(CH_3)_2$	B 1
95	CH_3	CH_3	NH	S	2-Cl, 3-Cl	B 1
96	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 1
97	CH_3	CH_3	NH	S	2-Cl, 5-Cl	B 1
98	CH_3	CH_3	NH	S	2-Cl, 6-Cl	B 1
99	CH_3	CH_3	NH	S	3-Cl, 4-Cl	B 1
100	CH_3	CH_3	NH	S	3-Cl, 5-Cl	B 1
101	CH_3	CH_3	NH	S	2-CH_3, 3-CH_3	B 1
102	CH_3	CH_3	NH	S	2-CH_3, 4-CH_3	B 1

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
103	CH_3	CH_3	NH	S	2-CH_3, 5-CH_3	B 1
104	CH_3	CH_3	NH	S	2-CH_3, 6-CH_3	B 1
105	CH_3	CH_3	NH	S	3-CH_3, 4-CH_3	B 1
106	CH_3	CH_3	NH	S	3-CH_3, 5-CH_3	B 1
107	CH_3	CH_3	NH	S	2-Cl, 3-CH_3	B 1
108	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 1
109	CH_3	CH_3	NH	S	2-Cl, 5-CH_3	B 1
110	CH_3	CH_3	NH	S	3-Cl, 4-CH_3	B 1
111	CH_3	CH_3	NH	S	3-Cl, 5-CH_3	B 1
112	CH_3	CH_3	NH	S	2-CH_3, 3-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
113	CH_3	CH_3	NH	S	2-CH_3, 4-Cl	B 1
114	CH_3	CH_3	NH	S	2-CH_3, 5-Cl	B 1
115	CH_3	CH_3	NH	S	2-F, 3-F	B 1
116	CH_3	CH_3	NH	S	2-F, 4-F	B 1
117	CH_3	CH_3	NH	S	2-F, 5-F	B 1
118	CH_3	CH_3	NH	S	2-F, 6-F	B 1
119	CH_3	CH_3	NH	S	2-F, 3-Cl	B 1
120	CH_3	CH_3	NH	S	2-F, 4-Cl	B 1
121	CH_3	CH_3	NH	S	2-F, 5-Cl	B 1
122	CH_3	CH_3	NH	S	2-F, 6-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
1 2 3	CH_3	CH_3	NH	S	2-F, 3-CH_3	B 1
1 2 4	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 1
1 2 5	CH_3	CH_3	NH	S	2-F, 3-Br	B 1
1 2 6	CH_3	CH_3	NH	S	2-F, 4-Br	B 1
1 2 7	CH_3	CH_3	NH	S	3-F, 4-Cl	B 1
1 2 8	CH_3	CH_3	NH	S	3-F, 5-Cl	B 1
1 2 9	CH_3	CH_3	NH	S	3-F, 4-CH_3	B 1
1 3 0	CH_3	CH_3	NH	S	3-F, 5-CH_3	B 1
1 3 1	CH_3	CH_3	NH	S	2-Br, 4-Cl	B 1
1 3 2	CH_3	CH_3	NH	S	2-Br, 4-Br	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
1 3 3	CH_3	CH_3	NH	S	2-Br, 4-CH_3	B 1
1 3 4	CH_3	CH_3	NH	S	3-Br, 4-Cl	B 1
1 3 5	CH_3	CH_3	NH	S	3-Br, 4-Br	B 1
1 3 6	CH_3	CH_3	NH	S	3-Br, 4-CH_3	B 1
1 3 7	CH_3	CH_3	NH	S	2-Cl, 4-Br	B 1
1 3 8	CH_3	CH_3	NH	S	3-Cl, 4-Br	B 1
1 3 9	CH_3	CH_3	NH	S	2-Cl, 4-I	B 1
1 4 0	CH_3	CH_3	NH	S	3-Cl, 4-I	B 1
1 4 1	CH_3	CH_3	NH	S	3-F, 4-Br	B 1
1 4 2	CH_3	CH_3	NH	S	3, 4-OCH_2O-	B 1

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
143	CH_3	CH_3	NH	S	2-CF_3, 3-Cl	B 1
144	CH_3	CH_3	NH	S	2-CF_3, 4-Cl	B 1
145	CH_3	CH_3	NH	S	2-CF_3, 3-Br	B 1
146	CH_3	CH_3	NH	S	2-CF_3, 4-Br	B 1
147	CH_3	CH_3	NH	S	2-F, 4-NO_2	B 1
148	CH_3	CH_3	NH	S	3-F, 4-NO_2	B 1
149	CH_3	CH_3	NH	S	2-Cl, 4-NO_2	B 1
150	CH_3	CH_3	NH	S	3-Cl, 4-NO_2	B 1
151	CH_3	CH_3	NH	S	2-F, 4-OCH_3	B 1
152	CH_3	CH_3	NH	S	3-F, 4-OCH_3	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
153	CH_3	CH_3	NH	S	2-Cl, 4-OCH_3	B 1
154	CH_3	CH_3	NH	S	3-Cl, 4-OCH_3	B 1
155	CH_3	CH_3	NH	S	2-OCH_3, 4-OCH_3	B 1
156	CH_3	CH_3	NH	S	3-OCH_3, 4-OCH_3	B 1
157	CH_3	CH_3	NH	S	2-Cl, 3-Cl, 4-Cl	B 1
158	CH_3	CH_3	NH	S	2-Cl, 4-Cl, 5-Cl	B 1
159	CH_3	CH_3	NH	S	2-Cl, 4-Cl, 6-Cl	B 1
160	CH_3	CH_3	NH	S	3-Cl, 4-Cl, 5-Cl	B 1
161	CH_3	CH_3	NH	S	2-Cl, 3-Cl, 4-CH_3	B 1
162	CH_3	CH_3	NH	S	2-Cl, 3-Cl, 4-Br	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
163	CH_3	CH_3	NH	S	2-Cl, 3-Cl, 4-I	B 1
164	CH_3	CH_3	NH	S	2-Cl, 4-Br, 6-Cl	B 1
165	CH_3	CH_3	NH	S	2-Cl, 4-CH_3, 6-Cl	B 1
166	CH_3	CH_3	NH	S	2-Br, 4-Cl, 6-Cl	B 1
167	CH_3	CH_3	NCHO	S	4-Cl	B 1
168	CH_3	CH_3	$NCOCH_3$	S	4-Cl	B 1
169	CH_3	CH_3	$NCOOCH_3$	S	4-Cl	B 1
170	CH_3	CH_3	$NCON(CH_3)_2$	S	4-Cl	B 1
171	CH_3	CH_3	NCONHPh	S	4-Cl	B 1
172	CH_3	CH_3	NCH_3	S	4-Cl	B 1

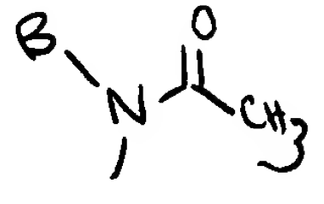

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
173	CH_3	CH_3	NC_2H_5	S	4-Cl	B 1
174	CH_3	CH_3	NC_3H_7	S	4-Cl	B 1
175	CH_3	CH_3	NCH_2OCH_3	S	4-Cl	B 1
176	CH_3	CH_3	$NCH_2CH_2OCH_3$	S	4-Cl	B 1
177	CH_3	CH_3	$NCH_2CH=CH_2$	S	4-Cl	B 1
178	CH_3	CH_3	$NCH_2C\equiv CH$	S	4-Cl	B 1
179	CH_3	CH_3	NCH_2COCH_3	S	4-Cl	B 1
180	CH_3	CH_3	NCH_2COOCH_3	S	4-Cl	B 1
181	CH_3	CH_3	$NCH_2COOC_2H_5$	S	4-Cl	B 1
182	CH_3	CH_3	NCH_2CN	S	4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
183	CH_3	CH_3	NCH_2Ph	S	4-Cl	B 1
184	CH_3	CH_3	$NCH_2C_6H_4$-4-Cl	S	4-Cl	B 1
185	CH_3	CH_3	$NCH_2C_6H_4$-4-CH_3	S	4-Cl	B 1
186	CH_3	CH_3	NCH_2CH_2Ph	S	4-Cl	B 1
187	CH_3	CH_3	NSO_2CH_3	S	4-Cl	B 1
188	CH_3	CH_3	$NSO_2N(CH_3)_2$	S	4-Cl	B 1
189	CH_3	CH_3	NCHO	S	4-Cl	B 2
190	CH_3	CH_3	NCH_3	S	4-Cl	B 2
191	CH_3	H	NH	S	4-Cl	B 1
192	CH_3	H	NH	S	2-Cl, 4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
193	CH_3	H	NH	S	3-Cl, 4-CH_3	B 1
194	CH_3	C_2H_5	NH	S	4-Cl	B 1
195	CH_3	C_2H_5	NH	S	4-Br	B 1
196	CH_3	C_2H_5	NH	S	2-Cl, 4-Cl	B 1
197	CH_3	C_2H_5	NH	S	2-Cl, 4-CH_3	B 1
198	CH_3	i-C_3H_7	NH	S	4-Cl	B 1
199	CH_3	i-C_3H_7	NH	S	4-Br	B 1
200	CH_3	i-C_3H_7	NH	S	2-Cl, 4-Cl	B 1
201	CH_3	i-C_3H_7	NH	S	2-Cl, 4-CH_3	B 1
202	CH_3	t-C_4H_9	NH	S	4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
203	CH_3	t-C_4H_9	NH	S	2-Cl, 4-Cl	B 1
204	CH_3	t-C_4H_9	NH	S	3-Cl, 4-CH_3	B 1
205	CH_3	CF_3	NH	S	4-Cl	B 1
206	CH_3	CF_3	NH	S	4-Br	B 1
207	CH_3	CF_3	NH	S	2-Cl, 4-Cl	B 1
208	CH_3	CF_3	NH	S	3-Cl, 4-CH_3	B 1
209	CH_3	H	NH	S	4-Cl	B 2
210	CH_3	H	NH	S	2-Cl, 4-Cl	B 2
211	CH_3	C_2H_5	NH	S	4-Cl	B 2
212	CH_3	i-C_3H_7	NH	S	4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
213	CH_3	t-C_4H_9	NH	S	4-Cl	B 2
214	CH_3	CF_3	NH	S	2-Cl, 4-Cl	B 2
215	CH_3	CH_2Ph	NH	S	4-Cl	B 1
216	CH_3	CH_2Ph	NH	S	2-Cl, 4-Cl	B 1
217	CH_3	CH_2Ph	NCHO	S	2-Cl, 4-Cl	B 1
218	CH_3	CHO	NH	S	4-Cl	B 1
219	CH_3	CHO	NCHO	S	4-Cl	B 1
220	CH_3	CHO	NH	S	2-Cl, 4-Cl	B 1
221	CH_3	CHO	NCHO	S	2-Cl, 4-Cl	B 1
222	CH_3	$COCH_3$	NH	S	4-Cl	B 1

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
223	CH_3	$COCH_3$	NH	S	4-Br	B 1
224	CH_3	$COCH_3$	NH	S	2-Cl, 4-Cl	B 1
225	CH_3	$COCH_3$	NCHO	S	2-Cl, 4-Cl	B 1
226	CH_3	SO_2CH_3	NH	S	4-Cl	B 1
227	CH_3	SO_2CH_3	NH	S	2-Cl, 4-Cl	B 1
228	CH_3	SO_2CH_3	NH	S	3-Cl, 4-CH_3	B 1
229	CH_3	$CONHCH_3$	NH	S	4-Cl	B 1
230	CH_3	$CONHCH_3$	NH	S	4-Br	B 1
231	CH_3	$CONHCH_3$	NH	S	2-Cl, 4-Cl	B 1
232	CH_3	$CONHCH_3$	NH	S	3-Cl, 4-CH_3	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
233	CH_3	CH_2Ph	NH	S	4-Cl	B 2
234	CH_3	CH_2Ph	NH	S	2-Cl, 4-Cl	B 2
235	CH_3	$COCH_3$	NH	S	4-Cl	B 2
236	CH_3	$CONHCH_3$	NH	S	4-Cl	B 2
237	CH_3	SO_2CH_3	NH	S	4-Cl	B 2
238	CH_3	SO_2CH_3	NH	S	2-Cl, 4-Cl	B 2
239	H	CH_3	NH	S	4-Cl	B 1
240	H	CH_3	NH	S	2-Cl, 4-Cl	B 1
241	H	CH_3	NH	S	3-Cl, 4-CH_3	B 1
242	CF_3	CH_3	NH	S	4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
243	CF_3	CH_3	NH	S	4-Br	B 1
244	CF_3	CH_3	NH	S	2-Cl, 4-Cl	B 1
245	CF_3	CH_3	NH	S	2-Cl, 4-CH_3	B 1
246	C_2H_5	CH_3	NH	S	4-Cl	B 1
247	C_2H_5	CH_3	NH	S	4-Br	B 1
248	C_2H_5	CH_3	NH	S	2-Cl, 4-Cl	B 1
249	C_2H_5	CH_3	NH	S	2-Cl, 4-CH_3	B 1
250	i-C_3H_7	CH_3	NH	S	4-Cl	B 1
251	i-C_3H_7	CH_3	NH	S	2-Cl, 4-Cl	B 1
252	i-C_3H_7	CH_3	NH	S	3-Cl, 4-CH_3	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
253	t-C_4H_9	CH_3	NH	S	4-Cl	B 1
254	t-C_4H_9	CH_3	NH	S	4-Br	B 1
255	t-C_4H_9	CH_3	NH	S	2-Cl, 4-Cl	B 1
256	t-C_4H_9	CH_3	NH	S	3-Cl, 4-CH_3	B 1
257	H	CH_3	NH	S	4-Cl	B 2
258	H	CH_3	NH	S	2-Cl, 4-Cl	B 2
259	C_2H_5	CH_3	NH	S	4-Cl	B 2
260	i-C_3H_7	CH_3	NH	S	4-Cl	B 2
261	CF_3	CH_3	NH	S	4-Cl	B 2
262	CF_3	CH_3	NH	S	2-Cl, 4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
263	Cl	CH_3	NH	S	4-Cl	B 1
264	Cl	CH_3	NH	S	2-Cl, 4-Cl	B 1
265	Cl	CH_3	NH	S	3-Cl, 4-CH_3	B 1
266	Cl	CH_3	NCHO	S	4-Cl	B 1
267	Cl	CH_3	NH	S	4-Br	B 1
268	Cl	CH_3	NCHO	S	2-Cl, 4-Cl	B 1
269	Cl	CH_3	NCHO	S	2-Cl, 4-CH_3	B 1
270	CH_3O	CH_3	NH	S	4-Cl	B 1
271	CH_3O	CH_3	NH	S	4-Br	B 1
272	CH_3O	CH_3	NH	S	2-Cl, 4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
278	CH_3O	CH_3	NH	S	2-Cl, 4-CH_3	B 1
274	CH_3O	CH_3	NCHO	S	4-Cl	B 1
275	CH_3O	CH_3	NCHO	S	2-Cl, 4-Cl	B 1
276	CH_3O	CH_3	NCHO	S	3-Cl, 4-CH_3	B 1
277	CH_3S	CH_3	NH	S	4-Cl	B 1
278	CH_3S	CH_3	NH	S	4-Br	B 1
279	CH_3S	CH_3	NH	S	2-Cl, 4-Cl	B 1
280	CH_3S	CH_3	NH	S	3-Cl, 4-CH_3	B 1
281	CH_3S	CH_3	NCHO	S	4-Cl	B 1
282	CH_3S	CH_3	NCHO	S	2-Cl, 4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
283	Cl	CH_3	NH	S	4-Cl	B 2
284	Cl	CH_3	NH	S	2-Cl, 4-Cl	B 2
285	CH_3O	CH_3	NH	S	2-Cl, 4-Cl	B 2
286	CH_3S	CH_3	NH	S	2-Cl, 4-Cl	B 2
287	CH_3	CH_3	SO	S	4-Cl	B 1
288	CH_3	CH_3	SO_2	S	4-Cl	B 1
289	CH_3	CH_3	NH	SO	4-Cl	B 1
290	CH_3	CH_3	NH	SO_2	4-Cl	B 1
291	CH_3	CH_3	NH	SO	2-Cl, 4-Cl	B 1
292	CH_3	CH_3	NH	SO_2	2-Cl, 4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
293	CH_3	CH_3	NH	SO	3-Cl, 4-CH_3	B 1
294	CH_3	CH_3	NH	SO_2	3-Cl, 4-CH_3	B 1
295	CH_3	CH_3	S	O	4-Cl	B 1
296	CH_3	CH_3	SO	O	4-Cl	B 1
297	CH_3	CH_3	SO_2	O	4-Cl	B 1
298	CH_3	CH_3	S	O	2-Cl, 4-Cl	B 1
299	CH_3	CH_3	SO	O	2-Cl, 4-Cl	B 1
300	CH_3	CH_3	SO_2	O	2-Cl, 4-Cl	B 1
301	CH_3	CH_3	NH	O	4-Cl	B 1
302	CH_3	CH_3	NH	O	4-CH_3	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
303	CH_3	CH_3	NH	O	2-Cl, 4-Cl	B 1
304	CH_3	CH_3	NH	O	2-Cl, 4-CH_3	B 1
305	CH_3	CH_3	NH	O	3-Cl, 4-CH_3	B 1
306	CH_3	CH_3	NH	O	2-F, 4-Cl	B 1
307	CH_3	CH_3	NCHO	O	4-Cl	B 1
308	CH_3	CH_3	NCH_3	O	4-Cl	B 1
309	CH_3	CH_3	NH	O	4-Cl	B 2
310	CH_3	CH_3	NH	O	2-Cl, 4-Cl	B 2
311	CH_3	CH_3	NH	S	4-F	B 2
312	CH_3	CH_3	NH	S	4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
313	CH_3	CH_3	NH	S	4-CH_3	B 2
314	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 2
315	CH_3	CH_3	NH	S	3-Cl, 4-Cl	B 2
316	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 2
317	CH_3	CH_3	NH	S	3-Cl, 4-CH_3	B 2
318	CH_3	CH_3	NH	S	2-F, 4-F	B 2
319	CH_3	CH_3	NH	S	2-F, 4-Cl	B 2
320	CH_3	CH_3	NH	S	2-F, 4-Br	B 2
321	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 2
322	CH_3	CH_3	NH	S	3-F, 4-CH_3	B 2

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
323	CH_3	CH_3	NCHO	S	4-Cl	B 2
324	CH_3	CH_3	NCHO	S	2-Cl, 4-Cl	B 2
325	CH_3	CH_3	NCHO	S	3-Cl, 4-CH_3	B 2
326	CH_3	CH_3	NH	S	4-Cl	B 3
327	CH_3	CH_3	NH	S	4-Br	B 3
328	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 3
329	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 3
330	CH_3	CH_3	NH	S	3-Cl, 4-CH_3	B 3
331	CH_3	CH_3	NH	S	4-Cl	B 4
332	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 4

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
333	CH_3	CH_3	NH	S	4-Cl	B 5
334	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 5
335	CH_3	CH_3	NH	S	4-Cl	B 6
336	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 6
337	CH_3	CH_3	NH	S	4-Cl	B 7
338	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 7
339	CH_3	CH_3	NH	S	4-Cl	B 8
340	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 8
341	CH_3	CH_3	NH	S	4-Cl	B 9
342	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 9

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
343	CH_3	CH_3	NH	S	4-Cl	B 10
344	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 10
345	CH_3	CH_3	NH	S	3-Cl, 4-Cl	B 10
346	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 10
347	CH_3	CH_3	NH	S	3-F, 4-CH_3	B 10
348	CH_3	CH_3	NH	S	2-F, 4-Cl	B 10
349	CH_3	CH_3	NH	S	4-Cl	B 11
350	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 11
351	CH_3	CH_3	NH	S	4-Cl	B 12
352	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 12

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W.	B
353	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 1 2
354	CH_3	CH_3	NH	S	3-F, 4-CH_3	B 1 2
355	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 1 2
356	CH_3	CH_3	NH	S	3-Cl, 4-CH_3	B 1 2
357	CH_3	CH_3	NCHO	S	4-Cl	B 1 2
358	CH_3	CH_3	NCHO	S	2-Cl, 4-Cl	B 1 2
359	CH_3	CH_3	NH	S	4-Cl	B 1 3
360	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 1 3
361	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 1 3

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
362	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 13
363	CH_3	CH_3	NH	S	4-Cl	B 14
364	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 14
365	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 14
366	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 14
367	CH_3	CH_3	NH	S	4-Cl	B 15
368	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 15
369	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 15
370	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 15
371	CH_3	CH_3	NH	S	4-Cl	B 16

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
372	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 16
373	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 16
374	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 16
375	CH_3	CH_3	NH	S	4-Cl	B 17
376	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 17
377	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 17
378	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 17
379	CH_3	CH_3	NH	S	4-Cl	B 18
380	CH_3	CH_3	NH	S	2-Cl, 4-Cl	B 18
381	CH_3	CH_3	NH	S	2-Cl, 4-CH_3	B 18

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
382	CH_3	CH_3	NH	S	2-F, 4-CH_3	B 18
383	CH_3	CH_3	NCHO	S	2-Cl, 4-Cl	B 1
384	CH_3	CH_3	NCHO	S	3-Cl, 4-Cl	B 1
385	CH_3	CH_3	NCHO	S	2-F, 4-Cl	B 1
386	CH_3	CH_3	NCHO	S	2-F, 4-CH_3	B 1
387	CH_3	CH_3	NCHO	S	3-F, 4-Cl	B 1
388	CH_3	CH_3	NCHO	S	3-F, 4-CH_3	B 1
389	CH_3	CH_3	NCHO	S	2-Cl, 4-CH_3	B 1
390	CH_3	CH_3	NCHO	S	3-Cl, 4-CH_3	B 1
391	CH_3	CH_3	NCHO	S	2-Cl, 3-Cl, 4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
392	CH_3	CH_3	NCHO	S	2-Cl, 4-Cl, 5-Cl	B 1
393	CH_3	CH_3	$NCOCH_3$	S	2-Cl, 4-Cl	B 1
394	CH_3	CH_3	$NCOCH_3$	S	3-Cl, 4-Cl	B 1
395	CH_3	CH_3	$NCOCH_3$	S	2-F, 4-CH_3	B 1
396	CH_3	CH_3	$NCOCH_3$	S	2-Cl, 3-Cl, 4-Cl	B 1
397	CH_3	CH_3	NCH_3	S	2-Cl, 4-Cl	B 1
398	CH_3	CH_3	NCH_3	S	3-Cl, 4-CH_3	B 1
399	CH_3	CH_3	NNO	S	2-Cl, 4-Cl	B 1
400	CH_3	CH_3	NNO	S	3-Cl, 4-CH_3	B 1
401	CH_3	CH_3	NNH_2	S	4-Cl	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
402	CH_3	CH_3	NNH_2	S	2-Cl, 4-Cl	B 1
403	CH_3	CH_3	NNH_2	S	3-Cl, 4-CH_3	B 1
404	CH_3	CH_3	NSO_2CH_3	S	2-Cl, 4-Cl	B 1
405	CH_3	CH_3	NSO_2CH_3	S	2-F, 4-CH_3	B 1
406	CH_3	CH_3	NSO_2CH_3	S	2-Cl, 3-Cl, 4-Cl	B 1
407	CH_3	CH_3	CH_2	S	4-Cl	B 1
408	CH_3	CH_3	CH_2	S	2-Cl, 4-Cl	B 1
409	CH_3	CH_3	CH_2	S	2-Cl, 4-CH_3	B 1
410	CH_3	CH_3	CH_2	S	2-F, 4-CH_3	B 1
411	CH_3	CH_3	CH(OH)	S	4-Cl	B 1

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Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
412	CH_3	CH_3	CH(OH)	S	2-Cl, 4-Cl	B 1
413	CH_3	CH_3	CH(OH)	S	2-Cl, 4-CH_3	B 1
414	CH_3	CH_3	CH(OH)	S	3-Cl, 4-CH_3	B 1
415	CH_3	CH_3	CH(OH)	S	2-F, 4-CH_3	B 1
416	CH_3	CH_3	CH(OH)	S	3-F, 4-CH_3	B 1
417	CH_3	CH_3	$CH(OCH_3)$	S	4-Cl	B 1
418	CH_3	CH_3	$CH(OCH_3)$	S	2-Cl, 4-Cl	B 1
419	CH_3	CH_3	$CH(OCOCH_3)$	S	4-Cl	B 1
420	CH_3	CH_3	$CH(OCOCH_3)$	S	2-Cl, 4-Cl	B 1
421	CH_3	CH_3	$CH(OCOCH_3)$	S	2-Cl, 4-CH_3	B 1

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
422	CH_3	CH_3	$CH(OCOCH_3)$	S	2-F, 4-CH_3	B 1
423	CH_3	CH_3	CH(F)	S	4-Cl	B 1
424	CH_3	CH_3	CH(F)	S	2-Cl, 4-Cl	B 1
425	CH_3	CH_3	CH(Cl)	S	4-Cl	B 1
426	CH_3	CH_3	CH(Cl)	S	2-Cl, 4-Cl	B 1
427	CH_3	CH_3	C=O	S	4-Cl	B 1
428	CH_3	CH_3	C=O	S	2-Cl, 4-Cl	B 1
429	CH_3	CH_3	C=O	S	2-Cl, 4-CH_3	B 1
430	CH_3	CH_3	C=O	S	2-F, 4-CH_3	B 1
431	CH_3	CH_3	CH_2	S	4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W.	B
432	CH_3	CH_3	CH_2	S	2-Cl, 4-Cl	B 2
433	CH_3	CH_3	CH_2	S	2-Cl, 4-CH_3	B 2
434	CH_3	CH_3	CH_2	S	2-F, 4-CH_3	B 2
435	CH_3	CH_3	CH(OH)	S	4-Cl	B 2
436	CH_3	CH_3	CH(OH)	S	2-Cl, 4-Cl	B 2
437	CH_3	CH_3	CH(OH)	S	2-Cl, 4-CH_3	B 2
438	CH_3	CH_3	CH(OH)	S	3-Cl, 4-CH_3	B 2
439	CH_3	CH_3	CH(OH)	S	2-F, 4-CH_3	B 2
440	CH_3	CH_3	CH(OH)	S	3-F, 4-CH_3	B 2
441	CH_3	CH_3	CH(OCH_3)	S	4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
442	CH_3	CH_3	$CH(OCH_3)$	S	2-Cl, 4-Cl	B 2
443	CH_3	CH_3	$CH(OCOCH_3)$	S	4-Cl	B 2
444	CH_3	CH_3	$CH(OCOCH_3)$	S	2-Cl, 4-Cl	B 2
445	CH_3	CH_3	$CH(OCOCH_3)$	S	2-Cl, 4-CH_3	B 2
446	CH_3	CH_3	$CH(OCOCH_3)$	S	2-F, 4-CH_3	B 2
447	CH_3	CH_3	CH(F)	S	4-Cl	B 2
448	CH_3	CH_3	CH(F)	S	2-Cl, 4-Cl	B 2
449	CH_3	CH_3	CH(Cl)	S	4-Cl	B 2
450	CH_3	CH_3	CH(Cl)	S	2-Cl, 4-Cl	B 2
451	CH_3	CH_3	C=O	S	4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
452	CH_3	CH_3	C=O	S	2-Cl, 4-Cl	B 2
453	CH_3	CH_3	C=O	S	2-Cl, 4-CH_3	B 2
454	CH_3	CH_3	C=O	S	2-F, 4-CH_3	B 2
455	CH_3	CH_3	C(CH_3)(OH)	S	4-Cl	B 2
456	CH_3	CH_3	C(CH_3)(OH)	S	2-Cl, 4-Cl	B 2
457	CH_3	CH_3	C(CH_3)($OCOCH_3$)	S	4-Cl	B 2
458	CH_3	CH_3	C(CH_3)($OCOCH_3$)	S	2-Cl, 4-Cl	B 2
459	CH_3	CH_3	C(CH_3)(F)	S	4-Cl	B 2
460	CH_3	CH_3	C(CH_3)(F)	S	2-Cl, 4-Cl	B 2
461	CH_3	CH_3	C(C_2H_5)(OH)	S	4-Cl	B 2

Table 1 (continued)

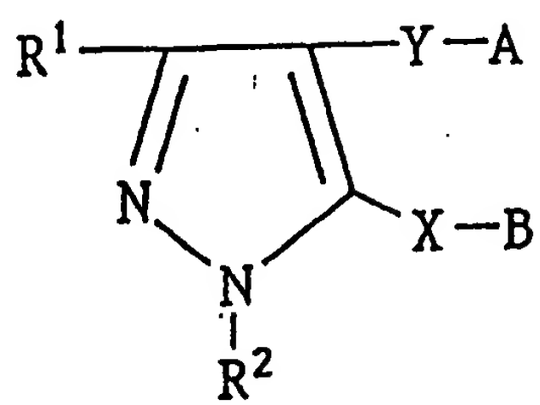
Compound No.	R^1	R^2	X	Y	W_n	B
462	CH_3	CH_3	$C(C_2H_5)(OH)$	S	2-Cl, 4-Cl	B 2
463	CH_3	CH_3	$C(i\text{-}C_3H_7)(OH)$	S	4-Cl	B 2
464	CH_3	CH_3	$C(i\text{-}C_3H_7)(OH)$	S	2-Cl, 4-Cl	B 2
465	CH_3	CH_3	$C(i\text{-}C_3H_7)(OH)$	S	2-Cl, 4-CH_3	B 2
466	CH_3	CH_3	$C(i\text{-}C_3H_7)(OH)$	S	2-F, 4-CH_3	B 2
467	CH_3	CH_3	$CH(CH_3)$	S	4-Cl	B 2
468	CH_3	CH_3	$CH(CH_3)$	S	2-Cl, 4-Cl	B 2
469	CH_3	CH_3	$CH(CH_3)$	S	2-Cl, 4-CH_3	B 2
470	CH_3	CH_3	$CH(CH_3)$	S	2-F, 4-CH_3	B 2
471	Cl	CH_3	CH_2	S	4-Cl	B 2

Table 1 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
472	Cl	CH_3	CH_2	S	2-Cl, 4-Cl	B 2
473	Cl	CH_3	CH_2	S	2-Cl, 4-CH_3	B 2
474	Cl	CH_3	CH_2	S	2-F, 4-CH_3	B 2
475	CH_3O	CH_3	CH_2	S	4-Cl	B 2
476	CH_3O	CH_3	CH_2	S	2-Cl, 4-Cl	B 2
477	CH_3O	CH_3	CH_2	S	2-Cl, 4-CH_3	B 2
478	CH_3O	CH_3	CH_2	S	2-F, 4-CH_3	B 2

Table 2

In compounds of:



Compound No.	R^1	R^2	X	Y	A	B
479	CH_3	CH_3	NH	S	A 1	B 1
480	CH_3	CH_3	NH	S	A 1	B 2
481	CH_3	CH_3	NH	S	A 2	B 1

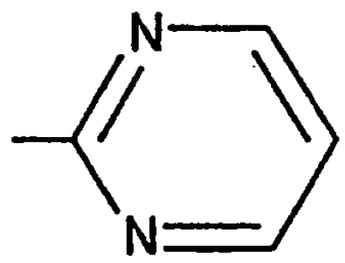
Table 2 (continued)

Compound No.	R^1	R^2	X	Y	W_n	B
482	CH_3	CH_3	NH	S	A 2	B 2
483	CH_3	CH_3	NH	S	A 3	B 1
484	CH_3	CH_3	NH	S	A 3	B 2
485	CH_3	CH_3	NH	S	A 4	B 1
486	CH_3	CH_3	NH	S	A 4	B 2
487	CH_3	CH_3	NH	S	A 5	B 1
488	CH_3	CH_3	NH	S	A 5	B 2
489	CH_3	CH_3	NH	S	A 6	B 1
490	CH_3	CH_3	NH	S	A 6	B 2
491	CH_3	CH_3	NH	S	A 7	B 1

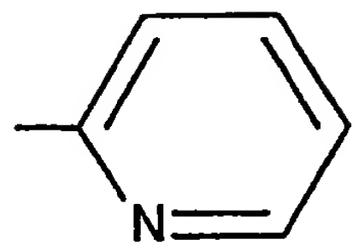
Table 2 (continued)

Compound No.	R^1	R^2	X	Y	W_a	B
492	CH_3	CH_3	NH	S	A 7	B 2
493	CH_3	CH_3	NH	S	A 8	B 1
494	CH_3	CH_3	NH	S	A 8	B 2
495	CH_3	CH_3	NH	S	A 9	B 1
496	CH_3	CH_3	NH	S	A 9	B 2
497	CH_3	CH_3	NH	S	A 10	B 1
498	CH_3	CH_3	NH	S	A 10	B 2
499	CH_3	CH_3	NH	S	A 11	B 1
500	CH_3	CH_3	NH	S	A 11	B 2

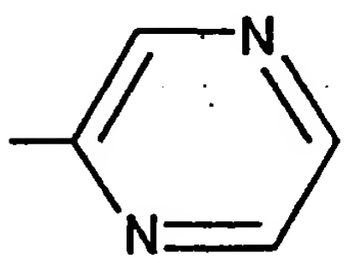
In the above-mentioned tables, B1 to B18 represent the following chemical structures:



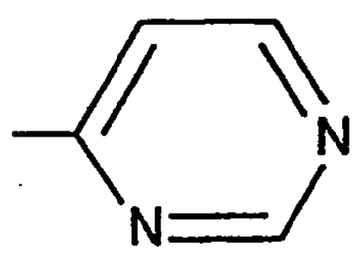
B1



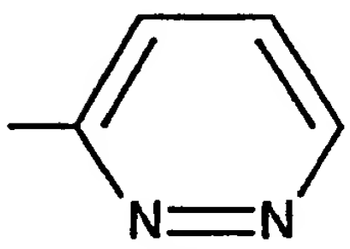
B2



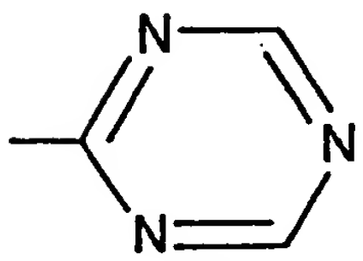
B3



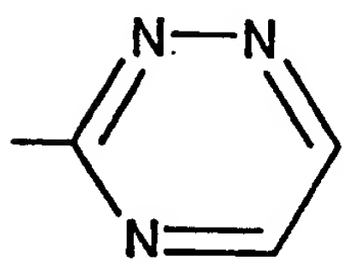
B4



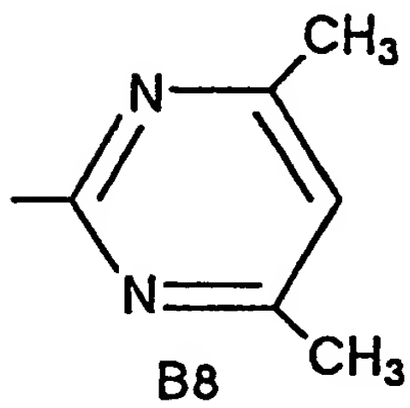
B5



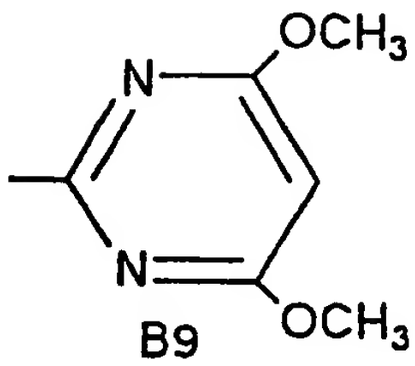
B6



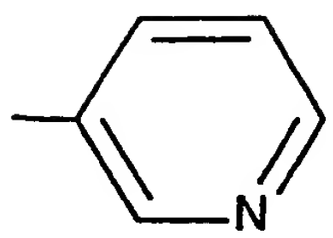
B7



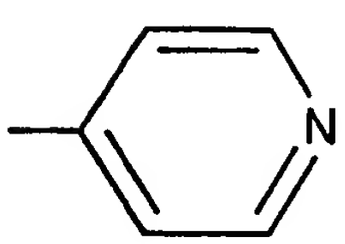
B8



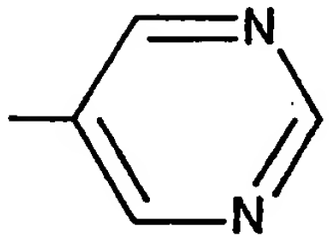
B9



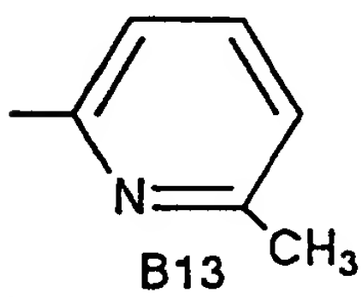
B10



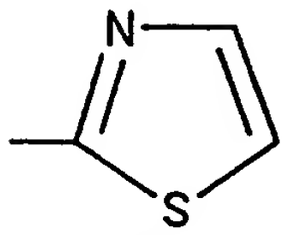
B11



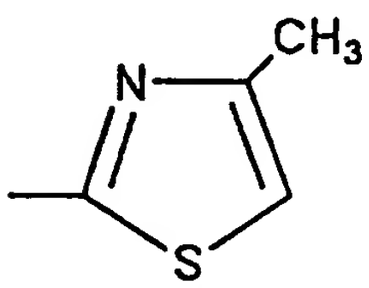
B12



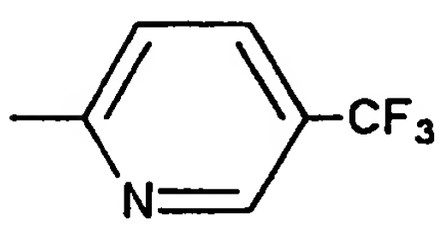
B13



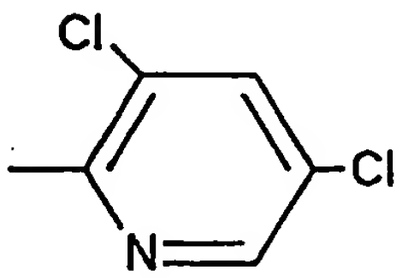
B14



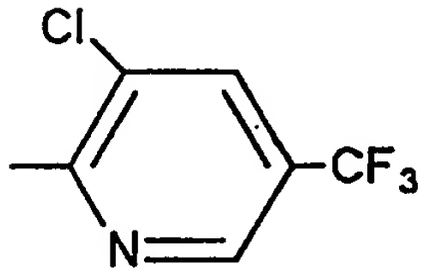
B15



B16

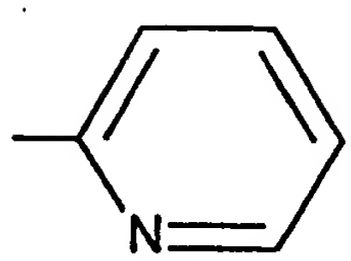


B17

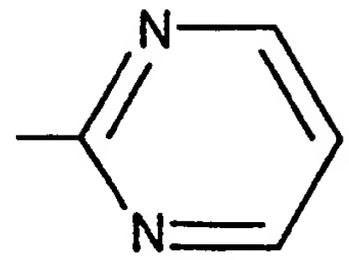


B18

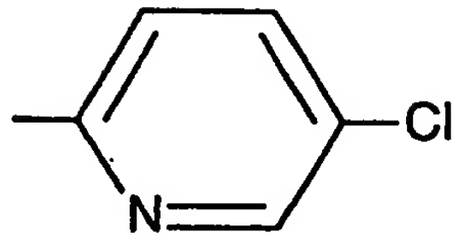
In the above-mentioned tables, A1 to A11 represent the following chemical structures:



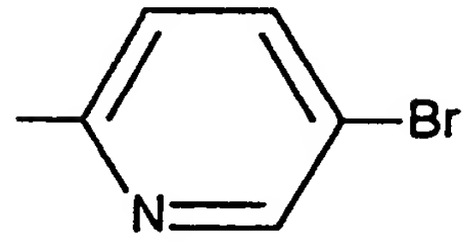
A1



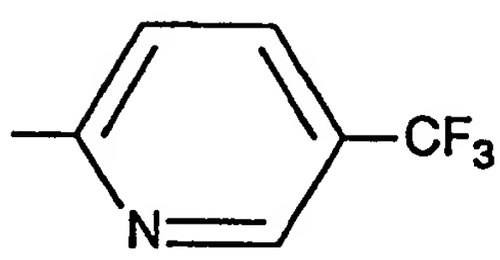
A2



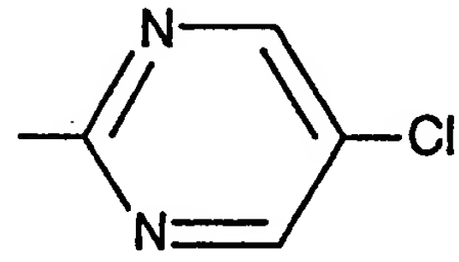
A3



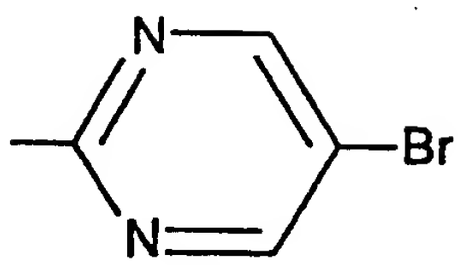
A4



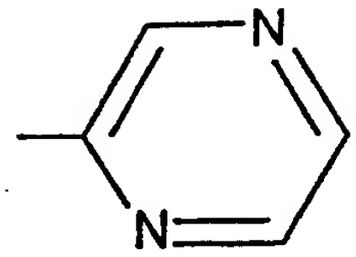
A5



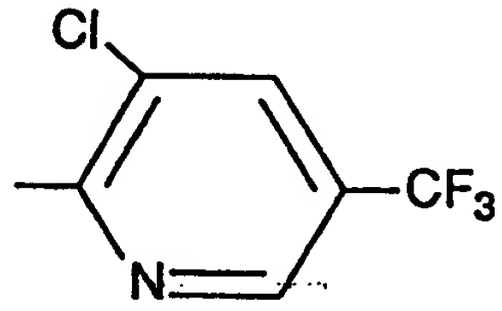
A6



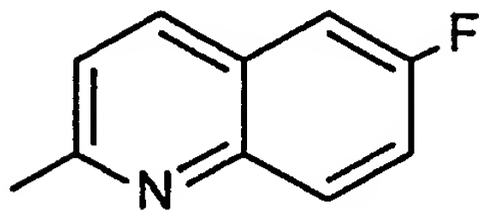
A7



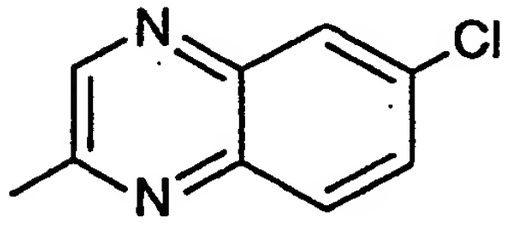
A8



A9



A10

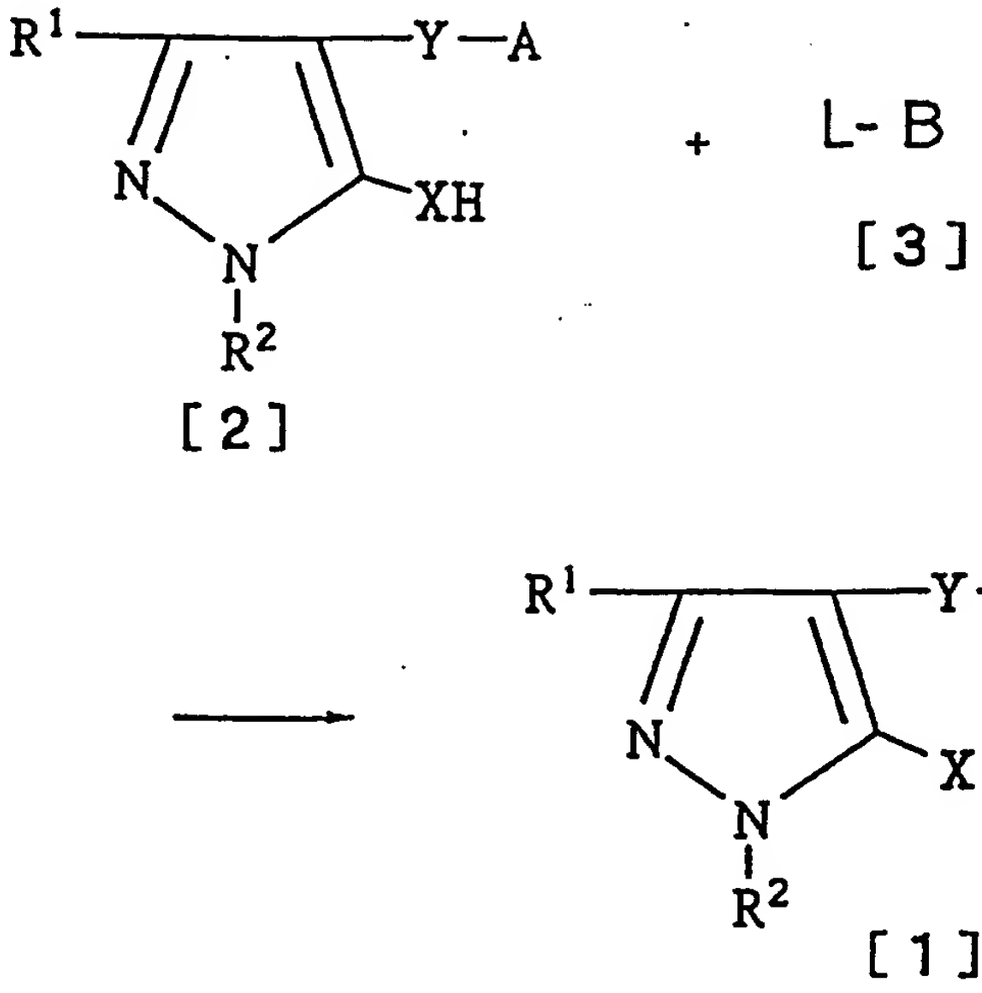


A11

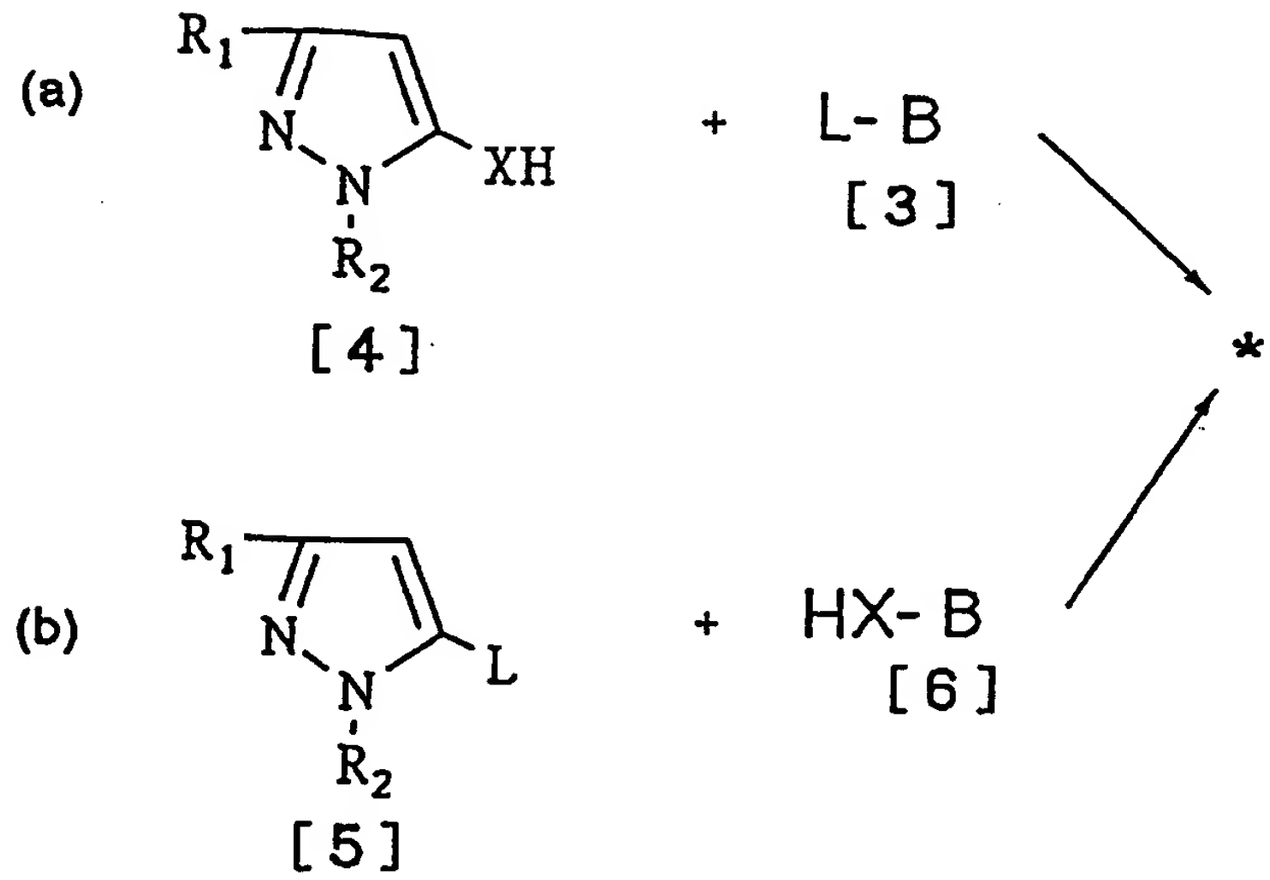
Next, methods of producing compounds of the present invention are shown below by way of the reaction schemes, which will be explained hereunder.

Reaction Schemes

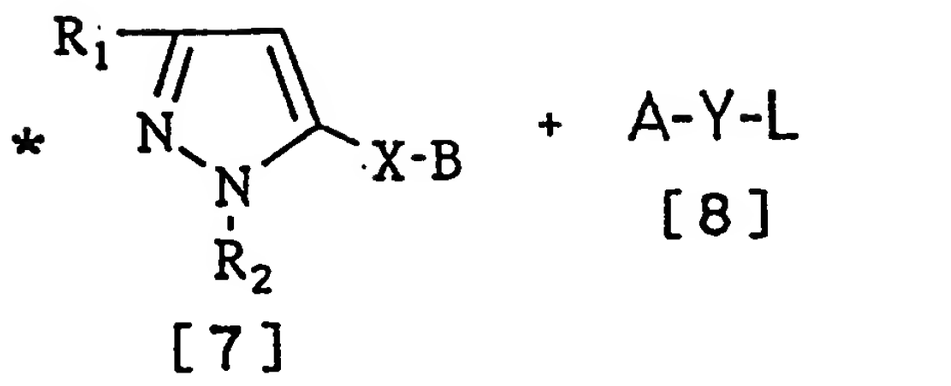
(Method 1)



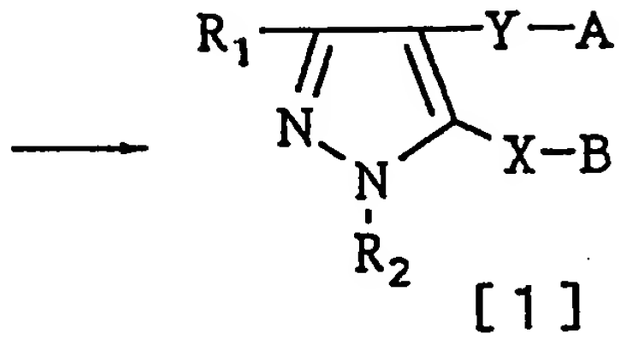
(Method 2)



When Y ≠ 0:



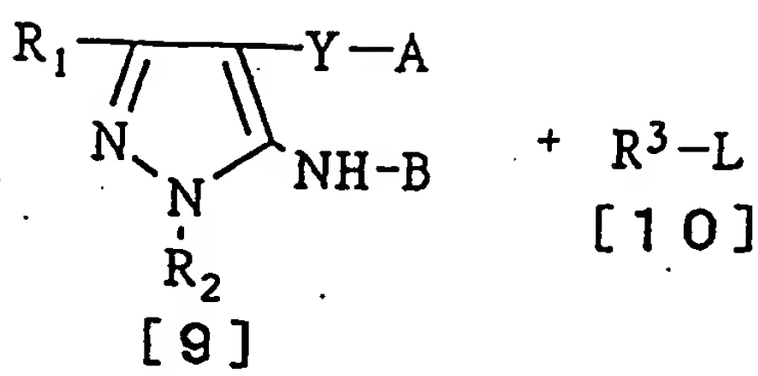
[7] [8]



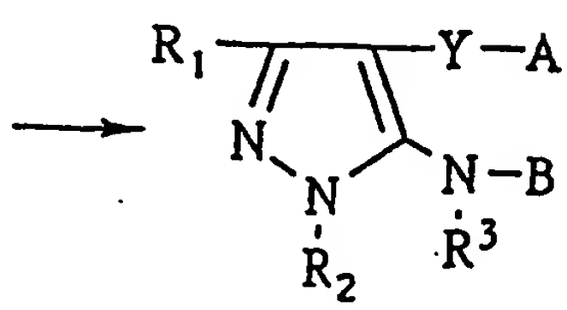
[1]

(Method 3)

When X = N-R³, R³ ≠ H:



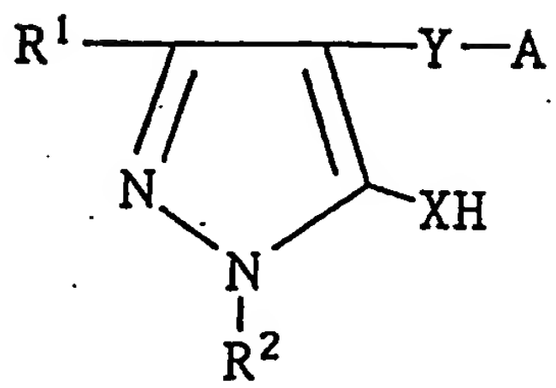
[9] [10]



[11]

(Method 1)

Compounds of the present invention are produced by reacting a substituted pyrazole of a general formula [2]:



[2]

where R^1, R^2, X, Y and A have the same meanings as mentioned above, and a heterocyclic compound of a general formula [3]:

L - B [3]

where L represents a leaving group such as a halogen atom or the like; and B has the same meaning as mentioned above. In the case, where X is $-NCOR^4$ or $-NSO_2R^5$, the product may be hydrolyzed by post-treatment or the like to give a compound where X is -NH.

The above-mentioned reaction does not always need a solvent. If needed, however, the solvent may selected from, for example, hydrocarbons such as toluene, xylene, chlorobenzene and the like, halogenated hydrocarbons such as dichloroethane and the like, ethers such as diisopropyl ether, dioxane and the like, esters such as ethyl acetate and the like, nitriles such as acetonitrile and the like, and polar solvents such as dimethylsulfoxide, dimethylformamide and the like.

If desired, an organic base (e.g., pyridine, triethylamine, etc.) or an inorganic base (e.g., potassium carbonate, sodium hydride, etc.) may be added to the reaction system.

If desired, a copper salt or a copper complex may also be added thereto as a catalyst.

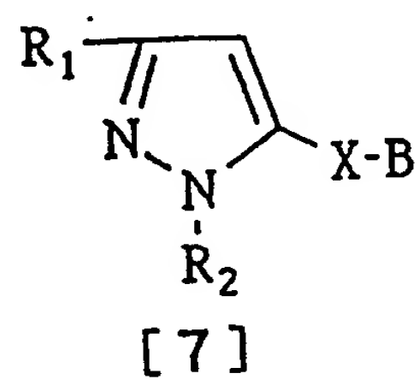
Regarding the amounts of the reactants of the above-mentioned reaction, the heterocyclic compound of formula [3] is within the range of from 1 to 5 equivalents to one equivalent of the substituted pyrazole of formula [2].

The reaction temperature of the reaction is not defined, but in general, it is preferably from room temperature to 200 °C or the reflux temperature of the solvent used.

After the reaction, the intended product may be obtained by ordinary treatment of the reaction mixture.

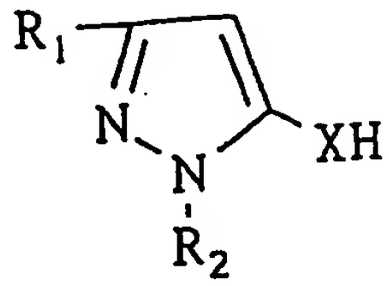
(Method 2)

(a) A compound of a general formula [7]:



[7]

where R^1,R^2,X and B have the same meanings as mentioned above, is produced by reacting a pyrazole of a general formula [4]:



[4]

where R^1, R^2 and X have the same meanings as mentioned above, and a heterocyclic compound of formula [3] optionally in the presence of a suitable solvent and a suitable base. In the case, where X is $-NCOR^4$ or $-NSO_2R^5$, the product may be hydrolyzed by post-treatment or the like to give a compound where X is -NH.

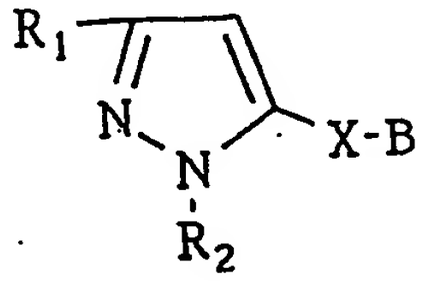
The solvent to be, if any, in the above-mentioned reaction includes, for example, hydrocarbons such as toluene, xylene, chlorobenzene and the like, halogenated hydrocarbons such as dichloroethane and the like, ethers such as diisopropyl ether, dioxane and the like, esters such as ethyl acetate and the like, nitriles such as acetonitrile and the like, and polar solvents such as dimethylsulfoxide, dimethylformamide and the like.

The base to be, if any, in the same includes, for example, potassium carbonate, sodium hydride and the like.

If desired, a copper salt or a copper complex may be added to the reaction system as a catalyst.

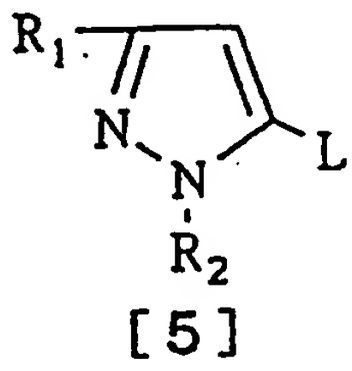
The reaction temperature of the reaction is not defined, but in general, it is preferably from room temperature to 200 °C or the reflux temperature of the solvent used.

(b) A compound of a general formula [7]:



[7]

where R^1, R^2, X and B have the same meanings as mentioned above is produced by reacting a pyrazole of a general formula [5]:



[5]

where R^1 and R^2 have the same meanings as mentioned above, and L represents a leaving group such as a halogen atom or the like, and a heterocyclic compound of a general formula [6]:

HX - B [6]

wher X and B hav th same meanings as mentioned above, opti nally in the pr sence of a suitable

solvent and a suitable base. In the case, where X is -NCOR^4 or -NSO$_2$R^5, the product may be hydrolyzed by post-treatment or the like to give a compound where X is -NH.

The solvent to be, if any, in the above-mentioned reaction includes, for example, hydrocarbons such as toluene, xylene, chlorobenzene and the like, halogenated hydrocarbons such as dichloroethane and the like, ethers such as diisopropyl ether, dioxane and the like, esters such as ethyl acetate and the like, nitriles such as acetonitrile and the like, and polar solvents such as dimethylsulfoxide, dimethylformamide and the like.

The base to be, if any, in the same includes, for example, potassium carbonate, sodium hydride and the like.

If desired, a copper salt or a copper complex may be added to the reaction system as a catalyst.

The reaction temperature of the reaction is not defined, but in general, it is preferably from room temperature to 200 • C or the reflux temperature of the solvent used.

Next, the pyrazole of formula [7] as obtained in the above-mentioned reaction (a) or (b) is reacted with a compound of a general formula [8]:

A - Y - L [8]

where A has the same meaning as mentioned above, Y has the same meaning as mentioned above, except oxygen atom, and L represents a leaving group such as a halogen atom or the like, optionally in the presence of a suitable solvent and a suitable base, to give a compound of the present invention.

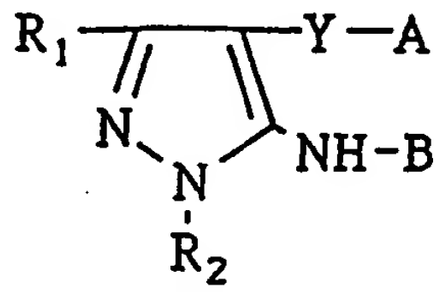
The solvent to be, if any, in the above-mentioned reaction includes, for example, hydrocarbons such as toluene, xylene, chlorobenzene and the like, halogenated hydrocarbons such as dichloroethane, chloroform, carbon tetrachloride and the like, ethers such as diisopropyl ether, dioxane and the like, esters such as ethyl acetate and the like, nitriles such as acetonitrile and the like, and polar solvents such as dimethylsulfoxide, dimethylformamide and the like.

The base to be, if any, in the same includes, for example, pyridine, triethylamine, potassium carbonate and the like.

The reaction temperature of the reaction is not defined, but in general, it is preferably from 0 • C to 100 • C.

(Method 3) When X = N-R^3, R^3 ≠ H:

A compound of the present invention is produced by reacting a pyrazole of a general formula [9]:



[9]

where R^1, R^2, Y, A and B have the same meanings as mentioned above, and a compound of a general formula [10]:

R^3 - L [10]

where R^3 has the same meaning as mentioned above except hydrogen atom, and L represents a leaving group such as a halogen atom or the like, optionally in the presence of a suitable solvent and a suitable base.

The solvent to be, if any, in the above-mentioned reaction includes, for example, hydrocarbons such as benzen , toluene, xylene and th lik , halogenated hydrocarbons such as dichloroethane, chloroform, carbon tetrachloride and the lik , ethers such as diisopropyl ether, tetrahydrofuran, dioxane and the like, esters such as ethyl acetat and th lik , nitriles such as acetonitril and the like, and polar solv nts such as dimethylsulfoxide, dimethylformamide and the lik .

The base to be, if any, in the same includes, for example, organic bases such as pyridin , triethylamine and the like, and inorganic bases such as potassium carbonate, sodium hydride and the like.

The reaction temperature of the reaction is not defined, but in general, it is preferably from 0 • C to 100 • C.

EXAMPLES

Next, concrete production examples are mentioned below.

Preparation Example 1 (preparation of compound No. 5 of the invention)

1.4 g of 4-(4-chlorophenylthio)-1,3-dimethyl-5-mercaptopyrazole and 1.2 g of 2-chloropyridine were stirred under heat at 120 • C for 1.5 hours. After cooled, 60 ml of ethyl acetate was added thereto and stirred, and the insoluble components were taken out by filtration. The filtrate was concentrated and then purified by silica gel column chromatography (developing solution; chloroform/ethyl acetate = 9/1), to give 0.6 g of 4-(4-chlorophenylthio)-1,3-dimethyl-5-(2-pyrimidylthio) pyrazole. Oily product. $n_D^{21.0}$ = 1.6465.

Preparation Example 2 (preparation of compound No. 49 of the invention)

① Preparation of N-(1,3-dimehtyl-5-pyrazolyl)formamide:

20 g of 5-amino-1,3-dimethylpyrazole was dissolved in 29 g of formic acid (85 %), and 55 g of acetic anhydride was dropwise added thereto under cooling with ice. After stirred for 3 days at room temperature, the reaction mixture was concentrated under reduced pressure and then purified by silica gel column chromatography (developing solution; chloroform) to give 12.8 g of N-(1,3-dimethyl-5-pyrazolyl)formamide.

② Preparation of 1,3-dimethyl-5-(2-pyridylamino)pyrazole:

A mixed solution comprising 4.1 g (29 mmol) of N-(1,3-dimethyl-5-pyrazolyl)formamide and 10 ml of N,N-dimethylformamide was dropwise added to a suspension of 70 ml of N,N-dimethylformamide containing 1.6 g of sodium hydride (55 %), under cooling with ice. After this was stirred for 2 hours at room temperature, a mixed solution comprising 3.4 g (30 mmol) of 2-chloropyrimidine and 10 ml of N,N-dimethylformamide was added thereto. After this was stirred under heat at 100 • C for further 2 days, the solvent was removed therefrom by distillation under reduced pressure and water was added thereto. Then, this was extracted with chloroform, washed with water and dried with anhydrous sodium sulfate. After this was filtered, the solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography to give 2.4 g of 1,3-dimethyl-5-(2-pyrimidylamino)pyrazole. m.p. 179.0 to 182.0 • C.

③ Preparation of compound No. 49 of the invention:

1.1 g of 1,3-dimethyl-5-(2-pyrimidylamino)pyrazole was dissolved in 30 ml of chloroform. 0.5 g of 4-chlorophenylsulfenyl chloride was dropwise added to the resulting solution at room temperature and reacted for one hour with stirring. The organic layer was washed with 30 ml of water and then dried with anhydrous sodium sulfate.

The solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography (developing solution; chloroform) to give 0.8 g of 4-(4-chlorophenylthio)-1,3-dimethyl-5-(2-pyrimidylamino)pyrazole. m.p. 158.0 to 159.0 • C.

Preparation Example 3 (preparation of compound No. 172 of the invention)

① Preparation of 1,3-dimethyl-5-(N-(2-pyrimidyl)-N-methylamino)pyrazole:

1.0 g of 1,3-dimethyl-5-(2-pyrimidylamino)pyrazole was added to a suspension of 10 ml of THF containing 0.25 g of sodium hydride (55 %), little by little under cooling with ice and then stirred for one hour at 60 • C. The solution was cooled to room temperature, and 4.1 g of methyl iodid was added thereto and refluxed gently for 2 hours. After cooled, 10 ml of water was add d thereto. Then, this was extracted three times each with 30 ml of diethyl ther. The ther layer was dried with anhydrous sodium sulfate, the

solvent was removed by distillation under reduced pressure, and the residu was purified by silica gel column chromatography (developing solution; chloroform) to give 0.5 g of 1,3-dimethyl-5-(N-(2-pyrimidyl)-N-methylamino)pyrazole. Yellow oily product.

② Preparation of compound No. 172 of the invention:

0.14 g of p-chlorophenylsulfenyl chloride was dropwise added to a solution of 10 ml of chloroform containing 0.2 g of 1,3-dimethyl-5-(N-(2-pyrimidyl)-N-methylamino)pyrazole and stirred for 15 hours at room temperature.

The solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography (developing solution; chloroform) to give 0.25 g of 4-(4-chlorophenylthio)-1,3-dimethyl-5-(N-(2-pyrimidyl)-N-methylamino)pyrazole. m.p. 77.0 to 78.0 °C.

Preparation Example 4 (preparation of compound No. 314 of the invention)

① Preparation of 1,3-dimethyl-5-(2-pyridylamino)pyrazole:

9.9 g of anhydrous potassium carbonate and 1 g of copper(II) acetylacetonate were added to a mixed solution of 60 ml of N,N-dimethylformamide containing 10 g of N-(1,3-dimethyl-5-pyrazolyl)formamide and 10.2 g of 2-bromopyridine and heated under reflux for 3 hours. After the solvent was removed by distillation under reduced pressure, water was added to the reaction mixture, which was then extracted with chloroform. The organic layer was washed with water and dried with anhydrous sodium sulfate.

The solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography (developing solution; chloroform/ethyl acetate) to give 4.6 g of 1,3-dimethyl-5-(2-pyridylamino)pyrazole. m.p. 113.0 to 115.0 °C.

② Preparation of compound No. 314 of the invention:

1.27 g of 1,3-dimethyl-5-(2-pyridylamino)pyrazole was dissolved in 50 ml of chloroform and cooled with ice water. 1.55 g of 2,4-dichlorophenylsulfenyl chloride was dropwise added to the solution and stirred for 15 hours at room temperature.

The solution was washed with an aqueous sodium hydrogencarbonate solution and then with water and dried with anhydrous sodium sulfate. The solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography (developing solution; chloroform) to give 1.75 g of 4-(2,4-dichlorophenylthio)-1,3-dimethyl-5-(2-pyridylamino)pyrazole. m.p. 144.0 to 145.0 °C.

Preparation Example 5 (preparation of compound No. 435 of the invention)

① Preparation of 2-pyridyl-(1,3-dimethyl-5-pyrazolyl)methanol:

300 ml of dry tetrahydrofuran solution containing 5.5 g of 2-bromopyridine was cooled to -78 °C, and 5.45 g of n-butyl lithium hexane solution (15 w/w %) was dropwise added thereto and stirred for 30 minutes. Then, 4.2 g of 1,3-dimethyl-5-formylpyrazole was dropwise added thereto. Afterwards, this was gradually heated up to room temperature and stirred for 15 hours.

The solution was neutralized by adding 2 N hydrochloric acid thereto, and then extracted three times each with 150 ml of ethyl acetate. The organic layer was dried with anhydrous sodium sulfate, the solvent was removed by distillation under reduced pressure, and the residue was purified by silica gel column chromatography (developing solution; chloroform) to give 5.2 g of 2-pyridyl-(1,3-dimethyl-5-pyrazolyl)-methanol as a brown oily product.

② Preparation of compound No. 435 of the invention:

3.4 g of p-chlorophenylsulfenyl chloride was dropwise added to 60 ml of dry chloroform solution containing 3 g of 2-pyridyl-(1,3-dimethyl-5-pyrazolyl)methanol at room temperature and stirred for 12 hours. After the solvent was rem ved by distillation, 50 ml of thyl acetat and 50 ml of aqueous 10 % sodium hydrogencarbonate solution were added to this and stirred for 30 minut s. The organic layer was s parated, and the aqueous layer was extracted three times each with 50 ml of ethyl acetate. The combined organic layers w r dried with anhydrous sodium sulfate. The solvent was removed by distillation und r reduced

pressur , and the r sidue was purified by silica g l column chromatography (developing solution; chloroform) to give 1.2 g of 2-pyridyl-4-(4-chloropehnylthio)-1,3-dimehtyl-5-pyrazolyl)methanol as white crystals. m.p. 90.0 to 91.0 °C.

Preparation Example 6 (preparation of compound No. 451 of the invention)

1.2 g of compound No. 435 of the invention as obtained in Production Example 5 was dissolved in 50 ml of dry dichloromethane, and 1.5 g of manganese oxide was added thereto at room temperature and stirred for 2 hours. The insoluble component was removed by filtration with Celite, the solvent was removed by distillation under reduced pressure, and the residue was crystallized from diisopropyl ether to give 1.0 g of 2-pyridyl-(4-(4-chlorophenylthio)-1,3-dimethyl-5-pyrazolyl)ketone as white crystals. m.p. 111.0 to 113.0 °C.

Physical properties of compounds as produced in accordance with these methods are shown in Table 3 below.

Table 3

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
5	oily $n_D^{21.0}$ = 1.6465	
47	m.p. 150.0~151.0 °C	
49	m.p. 158.0~159.0 °C	
52	m.p. 170.0~171.0 °C	
55	m.p. 150.0~152.0 °C	
56	m.p. 181.0~182.0 °C	
57	m.p. 190.0~193.0 °C	
58	m.p. 159.0~162.0 °C	

Table 3

Compound No	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
59	m.p. 168.5～171.5 ℃	
60	m.p. 157.0～158.0 ℃	
61	m.p. 170.5～172.5 ℃	
63	m.p. 160.0～162.0 ℃	
64	m.p. 156.0～157.0 ℃	
66	m.p. 180.0～183.0 ℃	
67	m.p. 125.5～127.0 ℃	
68	m.p. 154.0～157.0 ℃	
69	m.p. 126.0～127.0 ℃	

Table 3 (continued)

Compound No	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$) 、standard substance TMS
70	m.p. 178.0～179.0 ℃	
71	m.p. 163.5～167.5 ℃	
72	m.p. 174.0～176.0 ℃	
73	m.p. 132.5～135.5 ℃	
76	m.p. 148.0～151.0 ℃	
79	m.p. 127.5～128.5 ℃	
81	m.p. 180.0～184.0 ℃	
83	m.p. 175.0～177.0 ℃	

Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
95	m.p. 160.0～162.0 ℃	
96	m.p. 180.0～181.5 ℃	
97	m.p. 186.0～188.5 ℃	
98	m.p. 201.0～205.0 ℃	
99	m.p. 166.0～168.0 ℃	
100	m.p. 164.5～166.5 ℃	
102	m.p. 161.0～163.0 ℃	
105	m.p. 148.5～150.0 ℃	
106	m.p. 134.5～137.0 ℃	

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Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
108	m.p. 189.0～191.0 ℃	
110	m.p. 135.0～137.0 ℃	
113	m.p. 178.0～180.0 ℃	
116	m.p. 158.0～160.0 ℃	
120	m.p. 164.0～166.0 ℃	
124	m.p. 140.0～141.0 ℃	
126	m.p. 159.0～161.0 ℃	
129	m.p. 147.0～148.0 ℃	
132	m.p. 181.0～183.0 ℃	

Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
133	m.p. 196.0～198.0 ℃	
142	m.p. 162.0～164.0 ℃	
144	m.p. 143.0～145.0 ℃	
146	m.p. 151.5～153.5 ℃	
149	m.p. 214.0～216.0 ℃	
154	m.p. 139.0～140.0 ℃	
157	m.p. 192.5～195.5 ℃	
159	m.p. 190.5～193.5 ℃	
167	m.p. 123.0～124.0 ℃	

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Table 3 (continued)

Compound No	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
168	m.p. 152.0～154.0 ℃	
169	semi-crystalline	2.20(s,3H), 3.65(s,3H), 3.69(s,3H), 6.60～7.00(m,5H), 8.35(d,2H,J=5Hz)
170	m.p. 183.0～184.0 ℃	
172	m.p. 77.0～ 78.0 ℃	
175	m.p. 70.0～ 72.0 ℃	
176	oily	$n_D^{21.0}$ = 1.5957
177	m.p. 84.0～ 87.0 ℃	
178	m.p. 83.0～ 85.0 ℃	

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Table 3 (continued)

Compound No	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$) 、 standard substance TMS
179	oily	2.08(s,3H), 2.19(s,3H), 3.79(s,3H), 4.09(d,1H,J=18Hz), 5.10(d,1H,J=18Hz) 6.54(t,1H,J=5Hz), 6.90(s,4H), 8.13(d,2H,J=5Hz)
180	m.p. 98.0～100.0 ℃	
182	m.p. 128.0～129.0 ℃	
184	oily	2.20(s,3H), 3.21(s,3H), 4.50(d,1H,J=14Hz), 5.52(d,1H,J=14Hz), 6.58(t,1H,J=5Hz), 6.91(s,4H), 7.08(s,4H), 8.18(d,2H,J=5Hz)
187	m.p. 164.0～166.0 ℃	

Table 3 (continued)

Compound No	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$) 、 standard substance TMS
188	m.p. 189.0～191.0 ℃	
194	m.p. 142.0～143.5 ℃	
195	m.p. 140.5～141.5 ℃	
197	m.p. 149.0～152.0 ℃	
198	m.p. 168.5～169.5 ℃	
199	m.p. 164.5～167.0 ℃	
201	m.p. 183.0～187.0 ℃	
216	m.p. 205.0～207.0 ℃	
291	m.p. 194.0～196.0 ℃	

Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
292	m.p. 186.0～188.0 ℃	
301	m.p. 148.0～150.0 ℃	
303	m.p. 165.0～166.0 ℃	
311	m.p. 127.0～128.0 ℃	
312	m.p. 167.0～168.0 ℃	
314	m.p. 144.0～145.0 ℃	
315	m.p. 126.0～128.0 ℃	
316	m.p. 145.0～147.0 ℃	
317	m.p. 118.0～120.0 ℃	

Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
318	m.p. 112.0～114.0 ℃	
319	m.p. 99.0～100.0 ℃	
320	m.p. 99.0～100.0 ℃	
321	m.p. 122.0～124.0 ℃	
322	m.p. 107.0～108.0 ℃	
326	m.p. 158.0～160.0 ℃	
328	m.p. 168.0～169.0 ℃	
339	m.p. 147.0～148.0 ℃	
341	m.p. 128.0～130.0 ℃	

Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, CDCl$_3$) 、 standard substance TMS
343	m.p. 128.0～131.0 ℃	
344	m.p. 180.0～181.0 ℃	
345	m.p. 301.0～302.0 ℃	
346	m.p. 185.0～186.0 ℃	
347	m.p. 106.0～107.0 ℃	
351	m.p. 160.0～161.0 ℃	
352	m.p. 161.0～163.0 ℃	
354	m.p. 142.0～144.0 ℃	
355	m.p. 128.0～130.0 ℃	

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Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$) 、 standard substance TMS
363	m.p. 148.0～149.0 ℃	
365	m.p. 172.0～174.0 ℃	
367	m.p. 132.0～133.0 ℃	
391	m.p. 140.0～141.0 ℃	
402	resinous	2.15(s,3H), 3.70(s,3H), 4.62(bs,2H), 6.50～7.20(m,4H), 8.16(d,2H,J=5Hz)
435	m.p. 90.0～ 91.0 ℃	
436	m.p. 142.0～145.0 ℃	

Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
440	resinous	2.20(s,6H), 3.55(s,3H), 5.10(bs,1H), 6.05(s,1H), 6.50～7.60(m,6H), 8.40(d,1H,J=5Hz)
441	oily	2.17(s,3H), 3.30(s,3H), 3.68(s,3H), 5.68(s,1H), 6.80～7.70(m,7H), 8.30～8.50(m,1H)
443	oily	2.10(s,3H), 2.25(s,3H), 4.05(s,3H), 6.80～8.70(m,9H),

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Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
447	oily	2.19(s,3H), 3.77(s,3H), 6.67(d,1H,J=45Hz), 6.90～7.70(m,7H), 8.51(d,1H,J=5Hz)
451	m.p. 111.0～113.0 ℃	
455	oily	2.03(s,3H), 2.16(s,3H), 3.69(s,3H), 6.00(bs,1H), 6.80～8.51(m,8H)
463	resinous	0.50(d,3H,J=7Hz), 1.00(d,3H,J=7Hz), 2.02(s,3H), 3.21～3.65(m,1H), 3.87(s,3H), 6.00(bs,3H), 6.61～8.35(m,8H)

Table 3 (continued)

Compound No.	Physical properties	^{1}H-NMR δ (ppm, $CDCl_3$), standard substance TMS
481	semi-crystalline	2.26(s, 3H), 3.72(s, 3H), 6.62(t, 1H, J=5Hz), 6.88(t, 1H, J=5Hz), 7.61(bs, 1H), 8.23(d, 2H, J=5Hz), 8.34(d, 2H, J=5Hz)
483	m.p. 167.0～169.0 ℃	
485	m.p. 187.0～189.0 ℃	
497	m.p. 204.0～206.0 ℃	
499	m.p. 159.0～161.0 ℃	

Where compounds of the present invention are used as a fungicide for agricultural and horticultural use, in general, th y may be mixed with a suitable carri r, for example, a solid carrier such as clay, talc, bentonite, diatomaceous earth or the lik , or a liquid carrier such as water, alcohols (e.g., methanol, ethanol, etc.), aromatic hydrocarbons (.g., benz n , toluene, xylene, tc.), chlorinat d hydrocarbons, thers, ketones, esters (e.g., ethyl ac tate, etc.), acid, amides (e.g., dimethylformamid , etc.) or the like. If desired,

they may be blend d with an emulsifi r, a dispersing agent, a suspending agent, a penetrating agent, a spreader, a stabilizer and the like to be formed into various practical formulations of liquid, oil, emulsion, wettable powder, powder, granules, flowable or the like.

If desired, they may also be combined with any other herbicides, various insecticides, fungicides, plant growth regulators,synergists and others, in forming them into formulations or in actually sprinkling them onto plants.

The amount of the compound of the present invention to be applied to plants varies, depending upon the place, time, method, plant diseases, growing crops and other conditions. In general, the effective amount is suitably from 0.005 to 50 kg or so per ha (hectare).

Next, some examples of fungicidal formulations of containing the compound of the present invention as an active ingredient are shown below, which, however, are not limitative. In the following examples, "parts" are by weight.

Formulation Example 1: Emulsion

Compound of the invention	20 parts
Xylene	55 parts
N,N-dimethylformamide	20 parts
Sorpol 2680 (trade name by Toho Chemical Industry Co.; mixture of nonionic surfactant and anionic surfactant)	5 parts

The above ingredients are uniformly blended to form an emulsion. Before use, the emulsion is diluted to from 1/50 to 1/20000, and the diluted emulsion is sprayed over a crop field in an amount of from 0.005 to 50 kg, as the active ingredient, per ha.

Formulation Example 2: Wettable powder

Compound of the invention	25 parts
Zieklite PFP (trade name by Zieklite Industry Co.; mixture of kaolinite and sericite)	66 parts
Sorpol 5039 (trade name by Toho Chemical Industry Co.; anionic surfactant)	4 parts
Carplex #80 (trade name by Shionogi & Co., Ltd.; white carbon)	3 parts
Calcium lignin sulfonate	2 parts

The above ingredients are uniformly blended and milled to form a wettable powder. Before use, the powder is diluted with water to from 1/50 to 1/20000, and the diluted liquid is sprayed over a crop field in an amount of from 0.005 to 50 kg, as the active ingredient, per ha.

Formulation Example 3: Oil

Compound of the invention	10 parts
Methyl Cellosolve	90 parts

The above ingredients are uniformly blended to form an oil. For use, this is sprayed over a crop field in an amount of from 0.005 to 50 kg, as the active ingredient, per ha.

Formulation Example 4: Powder

Compound of the invention	3.0 parts
Carplex #80 (trade name by Shionogi & Co., Ltd.; white carbon)	0.5 parts
Clay	95 parts
Diisopropyl phosphate	1.5 parts

The above ingredients are uniformly blended and milled to form a powder. For use, this is sprayed over a crop filed in an amount of from 0.005 to 50 kg, as the active ingredient, per ha.

Formulation Example 5: Granules

Compound of the invention	5 parts
Bentonite	54 parts
Talc	40 parts
Calcium lignin sulfonate	1 part

The above ingredients are uniformly blended and milled , and a small amount of water is added thereto and mixed with stirring. The mixture is granulated through an granulating extruder and dried to form granules. For use, the granules are sprayed over a crop field in an amount of from 0.005 to 50 kg, as the active ingredient, per ha.

Formulation Example 6: Flowable

Compound of the invention	25 parts
Sorpol 3353 (trade name by Toho Chemical Industry Co.; nonionic surfactant)	10 parts
Lunox 1000C (trade name by Toho Chemical Industry Co.; anionic surfactant)	0.5 parts
1 % Zanthan gum aqueous solution (natural polymer)	20 parts
Water	44.5 parts

The above ingredients except the active ingredient are uniformly mixed , and the compound of the invention is added thereto and well stirred. The resulting blend is then wet-milled in a sand mill to obtain a flowable. Before use, this is diluted to from 1/50 to 1/20000, and the diluted liquid is sprayed over a crop field in an amount of from 0.005 to 50 kg, as the active ingredient, per ha.

Compounds of the present invention are effective for protecting plants from various plant diseases caused by, for example, Pyricularia oryzae, Cochliobolus miyabeanus, Rhizoctonia solani, Erysiphe graminis f. sp. hordei, f. sp. tritici, Pyrenophora graminea, Pyrenophora teres, Gibberella zeae, Puccinia striiformis, P. graminis, P. recondita, P. hordei, Typhula sp., Micronectriella nivais, Ustilago tritici, U. nuda, Pseudocercosporella herpotrichoides, Rhynchosporium secalis, Septoria tritici, Leptosphaeria nodorum, Dianorthe citri, Elsinoe fawcetti, Penicillium digitatum, P. italicum, Sclerotinia mali, Valsa mali, Podosphaera leucotricha, Alternaria mali, Venturia inaequalis, Venturia nashicola, Alternaria kikuchiana, Gymnosporangium haraeanum, Sclerotinia cinerea, Cladosporium carpophilum, Phomopsis sp., Plasmopara viticola, Elsinoe ampelina, Glomerella cingulata, Uncinula necator, Phakopsora ampelopsidis, Gloeosporium kaki, Cercospora kaki, Mycosphaerella nawae, Pseudoperenospora cubensis, Colletotrichum lagenarium, Sphaerotheca fuliginea, Mycosphaerella melonis, Phytophthora infestans, Alternaria solani, Cladosporium fulvam, Phomopsis vexans, Erysiphe cichoracoarum, Alternaria japonica, Cerocosporella brassicae, Puccinia allii, Cercospora kikuchii, Elsinoe glycines, Diaporthe phaseololum, Colletotrichum lindemuthianum, Mycosphaerella personatum, Cercospora arachidicola, Erysiphe pisi, Alternaria solani, Sphaerotheca humuli, Exobasidium r ticulatum, Elsinoe leucospila, Alternaria longipes, Erysiph cichoracearum, Colletotrichum tabacum, C rcospora beticola, Diplocarpon rosae, Sphaerotheca pannosa, Septoria chrysanthemiindici, Puccinia horiana, Botrytis cinerea, Sclerotinia sclerotiorum, etc.

The eff ctiveness of th compounds of the present invention is concr tely explain d by way of the following t st examples, which, how ver, are not limitative.

Test Example 1: Test for controlling gray mold (Botrytis cinerea)

An emulsion of the compound of the present invention was diluted with water to have a concentration of 500 ppm. This was sprayed over two- or three-leave-stage tomato plants (variety; Fukuju) as grown in a pot having a diameter of 7 cm with a spray gun, in an amount of 20 ml/pot.

On the next day, a suspension of spores of Botrytis cinerea (containing 1.0 % of glucose and 2.5 % yeast extract; 40 spores/visible area (× 150)) was sprayed over the plants, which were then put in a cultivation box having a temperature of 25 • C and a humidity of 95 % or more for 5 days. The region of the infected and spotted leaves to all the treated leaves was measured, and the preventive value of the compound was calculated out from the following equation.

Preventive value = [1-(region of infected leaves in treated group/region of infected leaves in control group)] x 100

As a result, the following compounds of the present invention showed the preventive value of being 100.

Compounds No. 5, NO. 49, No. 52, NO. 56, No. 58, No. 59, No. 63, No. 67, No. 68, No. 95, No. 96, No. 99, No. 105, No. 108, No. 110, No. 116, No. 120, No. 124, No. 126, No. 129, No. 132, No. 133, No. 142, No. 154, No. 157, No. 167, No. 172, No. 301, No. 311, No. 312, No. 314, No. 315, No. 316, No. 317, No. 318, No. 319, No. 320, No. 321, No. 322, No. 326, No. 328, No. 343, No. 383, No. 391, No. 402, No. 447.

Text Example 2: Test for controlling sheath blight (Rhizoctonia solani)

An emulsion of the compound of the present invention was diluted with water to have a concentration of 500 ppm. 5 ml/pot of this was applied to three- or four-leave-stage rice plant (variety; Nihonbare) as grown in a pot having a diameter of 5 cm, near the roots of them, and immediately thereafter, 15 ml/pot of this was sprayed over them.

Three days after the treatment, rice hulls as infected with Rhizoctonia solani were put near the roots of the plants and the plants were inoculated.

Then, the pots were put in a cultivation box having a temperature of 28 • C and a humidity of 95 % or more. Five days after the inoculation, the height of the infected and spotted leaves of the plants from the earth was measured, and the preventive value of the compound was calculated out from the following equation.

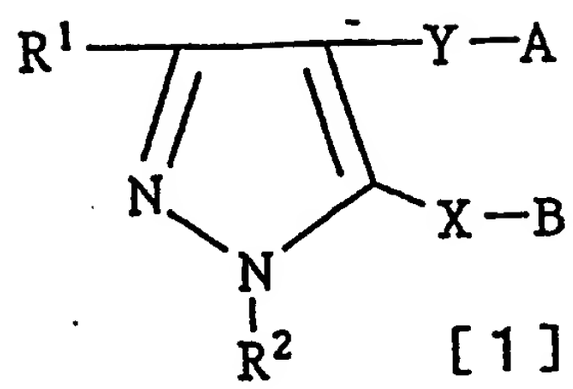
Preventive Value = [1-(height of infected leaves in treated group/height of infected leaves in control group)-] x 100

As a result, the following compounds of the present invention showed the preventive value of being 100.

Compounds No. 5, No. 47, No. 49, No. 52, No. 55, No. 56, No. 57, No. 59, No. 68, No. 69, No. 70, No. 71, No. 72, No. 79, No. 81, No. 96, No. 97, No. 99, No. 100, No. 102, No. 105, No. 108, No. 110, No. 113, No. 124, No. 126, No. 129, No. 142, No. 144, No. 146, No. 154, No. 157, No. 167, No. 168, No. 169, No. 172, No. 175, No. 178, No. 179, No. 182, No. 188, No. 194, No. 291, No. 292, No. 301, No. 312, No. 314, No. 316, No. 317, No. 318, No. 319, No. 320, No. 321, No. 322, No. 326, No. 328, No. 363, No. 365, No. 367, No. 383, No. 391, No. 402, No. 435, No. 436, No. 440, No. 441, No. 447, No. 451, No. 481, No. 483, No. 485, No. 499.

Claims

1. Substituted pyrazole derivatives of a general formula [1]:



[1]

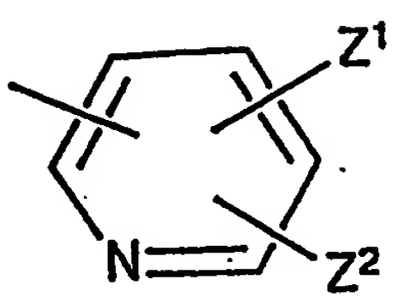
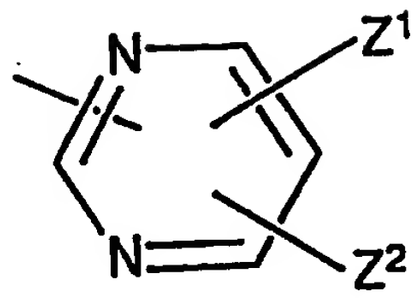
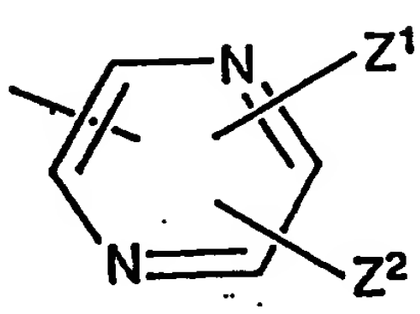
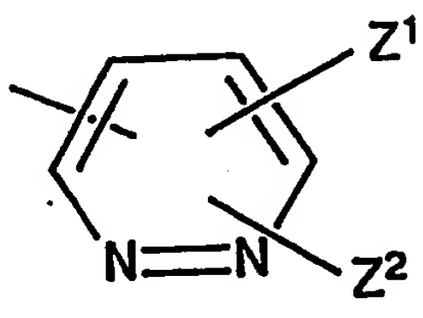
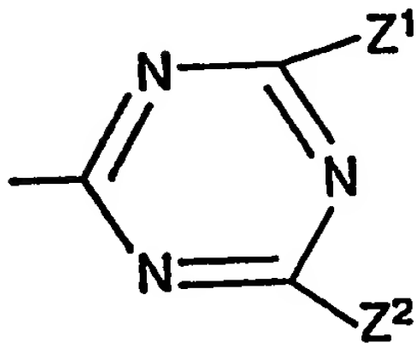
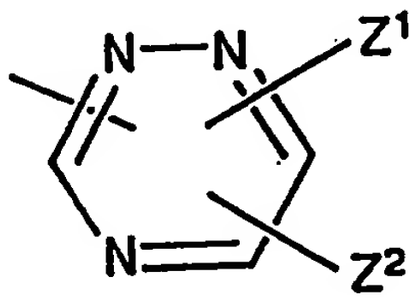
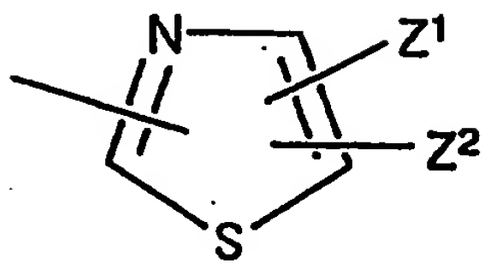
where R^1 represents a hydrogen atom, a halogen atom, an alkyl group, an alkoxy group, an alkylthio group or a haloalkyl group;
R^2 represents a hydrogen atom, an alkyl group, a haloalkyl group, an unsubstituted or substituted phenylalkyl group, $-COR^6$ or $-SO_2R^7$;
X represents -S-, -SO-, $-SO_2$-, $-N(R^3)$-, -CO- or $-C(R^4)(R^5)$-;
R^3 represents a hydrogen atom, an alkyl group, a haloalkyl group, an alkenyl group, an alkynyl group, an alkoxyalkyl group, a cyanoalkyl group, an alkylcarbonylalkyl group, an alkoxycarbonylalkyl group, a nitroso group, an amino group, an unsubstituted or substituted phenylalkyl group, $-COR^6$ or $-SO_2R^7$;
R^4 and R^5 independently represent a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkenyl group, an alkynyl group or $-OR^8$;
R^8 represents a hydrogen atom, an alkyl group, a haloalkyl group, an alkenyl group, an alkynyl group, an alkoxyalkyl group, a cyanoalkyl group, an alkylcarbonylalkyl group, an alkoxycarbonylalkyl group, an unsubstituted or substituted phenylalkyl group, $-COR^6$ or $-SO_2R^7$;
R^6 represents a hydrogen atom, an alkyl group, a haloalkyl group, an unsubstituted or substituted phenyl group, an unsubstituted or substituted phenylalkyl group, an alkoxy group or

$$-N<\begin{matrix}R^9\\R^{10}\end{matrix}$$;

R^7 represents an alkyl group, a haloalkyl group, an unsubstituted or substituted phenyl group or

$$-N<\begin{matrix}R^9\\R^{10}\end{matrix}$$;

R^9 and R^{10} independently represent a hydrogen atom, an alkyl group or an unsubstituted or substituted phenyl group;
Y represents an oxygen atom, -S-, -SO-, or $-SO_2$-;
A represents an unsubstituted or substituted phenyl group or an unsubstituted or substituted heterocyclic group;
B represents

or

;

Z¹ and Z² independently represent a hydrogen atom, a halogen atom, an alkyl group, an alkoxy group or a haloalkyl group.

2. Substituted pyrazole derivatives as claimed in claim 1, in which A is a substituted phenyl group.

3. Substituted pyrazole derivatives as claimed in claim 1, in which X is -N(R^3)-.

4. Substituted pyrazole derivatives as claimed in claim 2, in which Y is -S-.

5. Substituted pyrazole derivatives as claimed in claim 1, in which R^1 and R^2 each are a lower alkyl group, X is -N(R^3)-, Y is -S-, A is a substituted phenyl group, and B is an unsubstituted pyridyl group or an unsubstituted pyrimidyl group.

6. A fungicide for agricultural and horticultural use, containing one or mor substitut d pyrazoles as claimed in claim 1, as an active ingredi nt.

INTERNATIONAL SEARCH REPORT

International Application No PCT/JP91/01538

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

Int. Cl[5]

C07D401/06,401/12,401/14,403/06,403/12,403/14,417/06,417/12,
417/14,A01N43/54,43/56,43/58,43/60,43/707,43/78

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
IPC	C07D401/06-401/14, 403/06-403/14, 417/06-417/14, A01N43/54-43/62, 43/707, 43/78

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category *	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
A	JP, A, 01-125379 (Sumitomo Chemical Co., Ltd.), May 17, 1989 (17. 05. 89), (Family: none)	1-5, 6

* Special categories of cited documents: [10]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
January 8, 1992 (08. 01. 92)	January 28, 1992 (28. 01. 92)
International Searching Authority	**Signature of Authorized Officer**
Japanese Patent Office	